DEVELOPMENT OF COMPUTER CODE IN 'C' FOR INDEXING OF LAUE BACK REFLECTION PATTERNS FROM DIRECTIONALLY SOLIDIFIED Sn-Bi ALLOYS

A Thesis Submitted

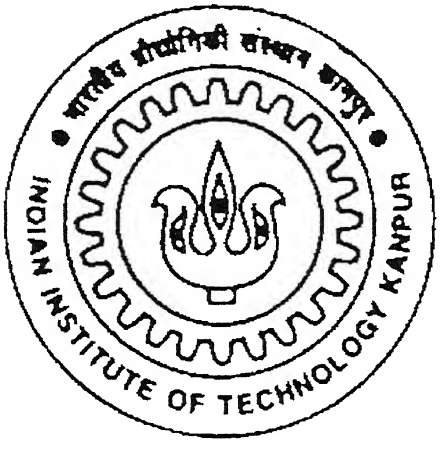
in Partial Fulfilment of the Requirements

for the Degree of

MASTER OF TECHNOLOGY

by

Ashish Ranjan



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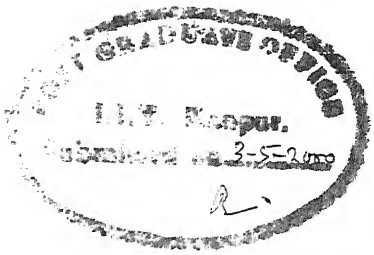

CERTIFICATE

This is to certify that the work contained in the thesis entitled "**Development of Computer code in 'C' for indexing of Laue back reflection patterns from directionally solidified Sn-Bi alloys**", by Mr. Ashish Ranjan has been carried out under my guidance and that this work has not been submitted elsewhere for a degree.

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At the end I wish to thank all those people who helped me directly or indirectly during my stay at IIT Kanpur.

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ABSTRACT

A computer code in 'C' language has been developed for indexing of Laue back reflection patterns from body centered tetragonal system. The developed code was used for indexing of two Laue patterns from directionally solidified Sn – 2 wt. % Bi alloys.

The sample used were from 11mm diameter ingots, which had been cut longitudinally through the ingot axis and also transversally. The transverse section was first prepared using conventional polishing technique followed by electropolishing. The electropolishing surface was exposed to white X-rays at 30 kV and 20mA for several hours to obtain Laue Back reflection patterns using Cu target.

The computer scheme is based on the conventional method of indexing. The indices are assigned to spots such that the interplaner angles between spots as well as the zone conditions are satisfied. The computer program assigns indices to spots and also determines the indices of important zone axes.

In the present work, attempts were made to verify the growth direction of primary dendrites in directionally solidified Sn-Bi alloys. The reported growth directions of primary dendrites were not confirmed for the alloys used in this study. The angular difference between the reported growth directions and the apparent growth direction in the samples has been found to vary in the range of 46 to 48 degrees when [1 1 0] was considered as the growth direction. When [1 $\bar{1}$ 0] was considered as growth direction the angular difference has been found to very in the range of 46 to 76. The angular difference between [0 0 1] and the ingot axis has been found to vary in the range of 40 to 58 degrees.

CHAPTER 1

INTRODUCTION

Determination of crystal orientation by Laue method involves indexing some of the spots and/or some of the zones of spots on the Laue photograph. Indexing of Laue for cubic crystals are generally easier, since the tables of interplanar angles and standard stereographic projections are available. When the crystal structure is non-cubic, indexing a pattern is then either very difficult or not possible. Since the labour involved in indexing Laue pattern is enormous, various approaches have been made to index the pattern by computer.

The present work involves developing a computer algorithm for the rapid indexing of Laue back reflection (LBR) patterns obtained from directionally solidified Sn - Bi alloys which have a body centered tetragonal structure. The algorithm has been written in 'C' language and the computer algorithm is based on the conventional method of matching interplanar angles between spots and also checking the zone condition. Reflections of planes having indices (6 6 6) to $(\bar{6}\,\bar{6}\,\bar{6})$ are indexed directly by using the x and y coordinates of the spots on the film. The input data for this program are the coordinates of spots on the pattern and the spot numbers belonging to one zone and zone angles measured from stereographic projection of the LBR pattern. The program matches the interplanar angles between spots and if the

difference between the experimental and theoretical angle lie within allowed error limit (± 3 degree) then the scheme checks the zone condition; i.e. spots belonging to one zone should satisfy the zone condition. The experimental angles are the angles calculated between spots using the coordinates of spots while theoretical angles are angles calculated from the crystallographic expression of body centered tetragonal system after assigning indices to the spots. The code developed finds out the indices for the important zone axes and the mutual angles between these axes are also matched with the corresponding angles obtained from the stereographic projection of the pattern.

The samples used in this work are Sn-Bi alloys, which belong to body centered tetragonal system, and have been directionally solidified. Directionally solidified alloys of tin had dendritic structure. The primary dendrites are plate shaped and are roughly parallel to the growth direction, which in turn is the ingot axis. It is noted from literature that, the growth direction in these alloys is [110], [1 $\bar{1}$ 0] and to a minor extent, [0 0 1]. This has been reported for pure tin as well as for Sn-12%Bi alloys [1, 2, 3, 4, 5]. The aim of this work is to compare the orientation reported in literature to the orientation obtained in this work. After indexing the spots the algorithm also assigns indices to zone axes from which the orientation of the crystal is determined and obtained orientation is matched with the orientation as described in literature.

Tin alloys with 2% bismuth were used for obtaining the Laue back reflection patterns for subsequent indexing. These alloys had been directionally solidified at different freezing rates. The directionally solidified ingot had been sectioned longitudinally as well as transversely. The transverse surface of the samples were ground and polished manually in the usual manner. Finally, the surface was electrolytically polished in a system, which has been developed as part of the present work. These samples are exposed to white radiation of X-rays at 20mA and 30 kV using cuk$_\alpha$ target and diffraction pattern is obtained for each sample. The input data is obtained from the pattern after making the stereographic projection for all the samples.

The computer program has been written in 'C' language and which assigns indices to spots as well to the zone axes. The orientation of the crystal can be easily determined after assigning indices to the spots and zone axes. This work involves development of a computer algorithm for indexing of LBR pattern for body centered tetragonal system and matching the measured orientation of directionally solidified Sn-Bi alloy to that published orientation in the literature.

CHAPTER 2

LITERATURE REVIEW

One of the convenient and accurate methods of determining the orientation of a single crystal or an individual grain in an aggregate is the Laue back-reflection method. In determining the orientation of a crystal from a pattern obtained from X-ray diffraction, it is necessary to assign the proper indices to some of the zones causing these hyperbolas or to some of the individual spots.

Indexing of Laue back reflection pattern can be tedious and time consuming, so attempts were made to index the pattern, using computer. The present literature review consists of work that has been carried out in this field by different workers.

One of the earlier attempts for indexing of Laue pattern by computer was carried out by D. T. Camp and J. A. Clum [6]. They produced a computer program for indexing of Laue back reflection pattern of zinc. They had written a digital – computer program which calculates and prints out a table of interplanar angles for an arbitrary crystal structure and then systematically searches through this table attempting to obtain a consistent fit to back reflection Laue pattern. The program was initially written in ALGOL and then a FORTRAN version was also produced. They used this scheme to index zinc, a hexagonal system. This program generates interplanar angles between indices (333) to ($\bar{3}\ \bar{3}\ \bar{3}$). The input data for their program are the crystal system to be indexed, parameters of that crystal system, angles between spots, which are measured by using stereographic projection. Then program compares the measured angles with the generated table of standard angles, and after checking the program returns indices of spots satisfying the condition.

W. H Huang, J. H. Christensen, and R. J. Black [7, 8] developed a computer algorithm for the rapid solution of Laue back reflection pattern of cubic crystal. Their algorithm duplicates the traditional technique of matching the angles between prominent reflections on the film with the true values of interplanar angles. The programs seeks a solution by assigning trial indices to a pair of reflections, generating a function of the true interplanar angle and comparing it with its counterpart from the X-ray pattern. They restricted their search to (333) to $(\bar{3}\ \bar{3}\ \bar{3})$, for indexing the spots. The uncertainties resulting from the spot size on the X-ray pattern and tilting of film were accommodated by permitting small allowable value of error for matching. The program was written in FORTRAN IV. The input data for their program are: specimen to film distance, the acceptable error in angles in matching, the number of reflections to be indexed, and the coordinates of spots in centimeters. The program first calculates and prints out the experimental angles between all pairs of reflections. It then begins the index search printing the successful trial values for each of the reflections. Final indices are assigned on the basis of angle matching among all the input points and are printed out, the root mean square error between the experimental angles and the true angles and prints best value.

C A Cornelius [9] produced a computer program for the simulation and analysis of X-ray back reflection Laue photographs of single crystal with any structure and orientation in FORTRAN IV. The program simulate both position and intensity of spots on the Laue back reflection X-ray photograph of a single crystal with any structure (including alloys) and of any orientation with respect to the X-ray beam. When Laue spots are indexed using computed patterns for orientation close to that of the specimen, another part of the program may be used to deduce the exact orientation.

An iterative program in FORTRAN IV has been written by J. Laugier [10] which allows the orientation matrix of a crystal to be determined and refined

without ambiguity, from a transmission or reflection Laue photograph. He designed the program to solve the Laue pattern without using the tedious input of a large number of data. The main Characteristics of his program are as follows. It is an iterative FORTRAN IV program needing a graphics screen, and optionally using a plotter. It caters for the seven crystal systems and measurements in both transmission and reflection. The parameters of the indexing procedure may be modified during successive trials. They considered only 11 Laue symmetry groups for getting only one solution for the pattern. The remaining solutions are, which are usually few, are stored in memory and the corresponding simulated Laue patterns are displayed. The input data for their program are the observed (x, y) coordinates of N Bragg spots on the Laue photographs. The indexing procedure starts with two reflections, which are at the intersection of the zonal planes and thus have small Miller indices. The angle between the corresponding scattering vectors is compared, within a given angular tolerance, to the angle between all possible pairs of reciprocal vectors up to a given limit for the Miller indices. The program is written in such a way that it rejects the equivalent solutions. This is done during the first step of the indexing procedure. Each time a given pair of reciprocal vectors satisfies the angular test, the operation of the Laue symmetry of the crystal are applied, thus generating a set of equilateral pairs of vector. These pairs are then compared to the pairs of vectors already selected and are rejected if they are identical.

Haskel V Hart [11] have developed a computer method, which may be used for indexing of symmetrical or asymmetrical transmission or back reflection Laue patterns of any crystal. They used their program for indexing Laue back reflection pattern of echinoid calcite having cubic structure. The input data for their program is either coordinates of spots read from film or Greninger angles (γ, δ) for each spot. They have written the program in BASIC.

Ryoji ohba [12] proposed a method that applies pattern recognition techniques to the computer-aided indexing of X-ray diffraction spots. The indices of a set of spots are determined based on an analysis of the set of mutual angles between every pair of reciprocal lattice vectors corresponding to those spots. They applied this method to the automatic determination of the crystallographic orientation of an ice crystal. The basic principle of the method is to select reciprocal lattice vector for a spot so as to maximize the probability of coincidence of the measured angles between any pair of diffraction spots with those calculated from theoretically allowable reciprocal lattice vectors.

P. Sampat Kumar and V. Bansal [13] have developed a scheme of indexing of Laue back reflection pattern of body centered tetragonal system. They proposed an iterative scheme of matching the interplanar angles between the planes causing the spots. The interplanar angles are calculated using the x and y coordinates of the spots. The experimental angles and theoretical angles are compared. The spots are assigned Miller indices such that the differences between these two remain within an allowable error range. In addition to matching interplanar angles between spots the condition that certain spots should belong to same zone is also checked. Mutual angles between zone axes are also matched with the experimental angle (obtained from stereographic projection). They restricted the index search from (666) to $(\bar{6}\ \bar{6}\ \bar{6})$. The program was written in FORTRAN 77 and it indexes the spots of the Laue pattern and their zone axes.

Directionally solidified alloys of tin and bismuth have dendritic structure. To determine the orientation by Laue back reflection method, samples of tin-bismuth are exposed to X-ray of white radiation. The shortest wavelength for tin with absorption edge[14] are given below in the next page.

Element	kα2	kα1	kβ	kβ2	kβII	kβ	K Absorption edge
Sn	0.495053	0.490599	0.435877	0.435236	0.425915		0.42467

To get diffraction pattern from tin a suitable wavelength is required. If wavelength is less than that of absorption edge, the large absorption will cause the reflection to be weak. Moreover, the fluorescent radiation emitted from the specimen will produce a heavy background. The resulting pattern will not be clear, and it may be impossible to interpret them.

The pattern will improve as the gap between the absorption edge of the specimen and the wavelength of the incident radiation increases [15]. The appearance of a Laue photograph of a given crystal set up is considerably affected by a change of potential on the X-ray tube. If a low potential is used, the minimum wavelength is relatively large and the clear space near the center of the photograph is correspondingly large. The pattern is also much less rich in spots than when a high potential is applied. When minimum wavelength is large the spectral range from which wavelengths are available for reflection is more restricted than when minimum wavelength is small. The Laue photograph becomes richer in spots and these appear closer to the center as minimum wavelength is progressively reduced.

For Sn

$\lambda_{min} = 12.4/V$, where wavelength is in angstrom and voltage in kilovolt.

Absorption edge for Sn is 0.42467, so the minimum voltage of X-ray required for diffraction will be given by:

$0.42467 = 12.4 / V$

Hence,

$$V = 12.4 / 0.42467$$

$$= 29.15(KV)$$

The present work also involves determining orientation of Sn-Bi alloys and comparing it with the published data. G. F. Bolling, J. J. Kramer, and W. A. Tiller [1] studied the growth direction of Sn-Bi system. They studied a 99.999 % pure tin and found that there is a preference direction for grain growth in the metal. Grains grow in [1 1 0] direction preferentially, which is the dendrite growth direction.

Dendritc growth in metals was studied by F. Weinberg and B. Chalmers [2]. They studied the dendrite growth in 99.98% pure single crystal of Tin. The c crystallographic axis is perpendicular to the general direction of the solidification and is in a horizontal plane, i.e., parallel to the top surface of the crystal. The crystallographic axes a and b are at 45 degree to this horizontal plane. They shown that a number of dendrite rows grow parallel to the direction of solidification and have large vertical secondary branches.

Other Tin crystals which were grown, in which the c-axes was vertical and with a and b axes at 45 degree to the direction of solidification. In this case, vertical secondary branches were not observed. They concluded that in tin the dendrite branches grow predominantly in the [1 1 0] and [1 $\bar{1}$ 0] directions with very slight growth in the [001] direction.

P. J. Aheran and M. C. Flemings [3] studied the dendrite morphology of unidirectionally solidified Sn-Bi alloy with the help of X-ray. They found that columnar growth direction of primary arms in this alloy is [110] while secondary arms grow in the [1$\bar{1}\bar{1}$], [$\bar{1}$1$\bar{1}$], and [112] directions. They also found that

interstices between dendrite arms tend to fill in preferentially to form planes parallel to the heat flow direction.

They found that in Sn-Bi alloy, the direction of growth of primary dendrite arms is [110]; this corresponds to the major axis of columnar grains in unidirectional solidification, and is therefore the "columnar growth direction". Secondary arms grow in the [1 $\bar{1}$ $\bar{1}$] and [$\bar{1}$ 1$\bar{1}$] directions; these are perpendicular to the [110] direction and are 138 degree apart. Secondary arms in <111> directions were found only on one side of the primary arm; that is, secondary arms did not grow in the [$\bar{1}$ 1 1], [1 $\bar{1}$ 1], or [111] directions.

The experiments of Weinberg and Chalmers were repeated by S.O'Hara to determine whether the tin dendrite grow freely in a bulk melt with the same orientation as the pulled dendrites. He found with the help of X-ray diffraction results that the single crystal that grew from the dendritic seed had the same orientation. These results verify the growth direction of bulk tin dendrites as being 12 degree off the [110] towards the [001] direction.

S.O'Hara [4] studied the morphology of controlled growth of tin dendrites. Dendrites of tin were pulled from a supercooled melt. He determined the growth morphology of tin dendrites and the growth direction was found to be 12 degree away from the [110] direction towards the [001] direction.

J. C. Warner and J. D. Verhoeven [5] studied the morphology of tin dendrites in near-eutectic alloys using tin-rich Sn-Ag alloys for their study. They used Laue back reflection method for determination of orientation. Using LBR technique they found that dendrites grow preferentially in [1 1 0] direction and the sheets were found to lie in the [1 $\bar{1}$ 0] direction.

The computational scheme involves calculation of theoretical interplanar and interzonal angles. These values have been checked with published data for tin crystals by R. E. Troounfelker et al. [16] and B. S. Chandrashekhar et al. [17].

The lattice constant c and a are obtained from the work published by J. A. Lee et al. [18]. Lee et al. reported a change in c/a ratio from 0.5455 for pure tin to 0.5451 for 6%Bi in Tin. The effect of such a variable in c/a ratio on an interplanar angles were found to be low so c/a ratio for the calculation of interplanr angles in this work, has been used as 0.55.

CHAPTER 3

EXPERIMENTAL SETUP AND PROCEDURE

3.1 INTRODUCTION

The basic aim of experimental work in this project was to get input data for computer program. The experimental work can be divided into three sections:

a) Sample and sample preparation for X-ray exposure
b) Exposing sample to X-ray to obtain Laue pattern
c) Obtaining input data for computer program from X-ray pattern.

3.2 SAMPLE AND SAMPLE PREPARATION

In this work directionally solidified alloy of Tin-Bismuth were used for X-ray exposure. These samples were directionally solidified by Mr. Ravi Kumar [19]. Directionally solidified alloys of Tin-Bismuth have dendritic structure, and primary dendrites are of plate type. The Bismuth percentage was 2% by weight in tin. The ingots were solidified at different freezing rates.

These ingots were cut into two halves and small lengths. The length of samples varied from 2 to 3 cm while diameter of the ingots was approx. 11 mm. The shape of a sample is as given below

Arrow direction shows the direction of growth of crystal. The transverse section, which is shown, as hatched, was prepared for X-ray exposure. Arrow marks helps in determining the face that is to be polished for X-ray exposure.

Sample preparation for X-ray exposure involves polishing and etching of the transverse section. Samples were first polished on emery paper starting from 1/0 and go up to 4/0 sequentially. After polishing on the 4/0 emery paper samples were polished on wheel polisher using Al_2O_3 as polishing medium. The samples were etched and polished alternatively and microstructure was observed at each stage on microscope. The etchant used is 1 gm of $K_2Cr_2O_7$ + 6-7 ml of conc. HCl + 86 ml of distill water.

Since the alloys of Sn-Bi are very soft, mechanical polishing results in scratches, so it was decided to electropolish the sample on self fabricated electrolytic polishing setup.

The electrolytic polishing setup, which was designed and fabricated for this work, is briefly described here.

3.2.1 Electrolytic Polishing Setup:

The drawing of the electopolishing set up is shown in the figure 3.1. An L-shaped cathode made of tin as shown in figure 3.1 was used which was partly covered with glass to hold a tin cathode. It was connected to negative terminal of constant current constant voltage (cc-cv) source. Anode is sample, which was held in a Teflon holder. The figure of sample holder is shown in figure 3.2. Sample holder is made up of Teflon. It holds the sample and it has an arrangement to hold the sample tightly during the electrolytic polishing. Drawing of Teflon screw and Teflon supports which keeps sample tight are shown in figure 3.3. .
A brass tube holds the Teflon holder. The arrangement for holding the Teflon holder is shown if figure 3.4.

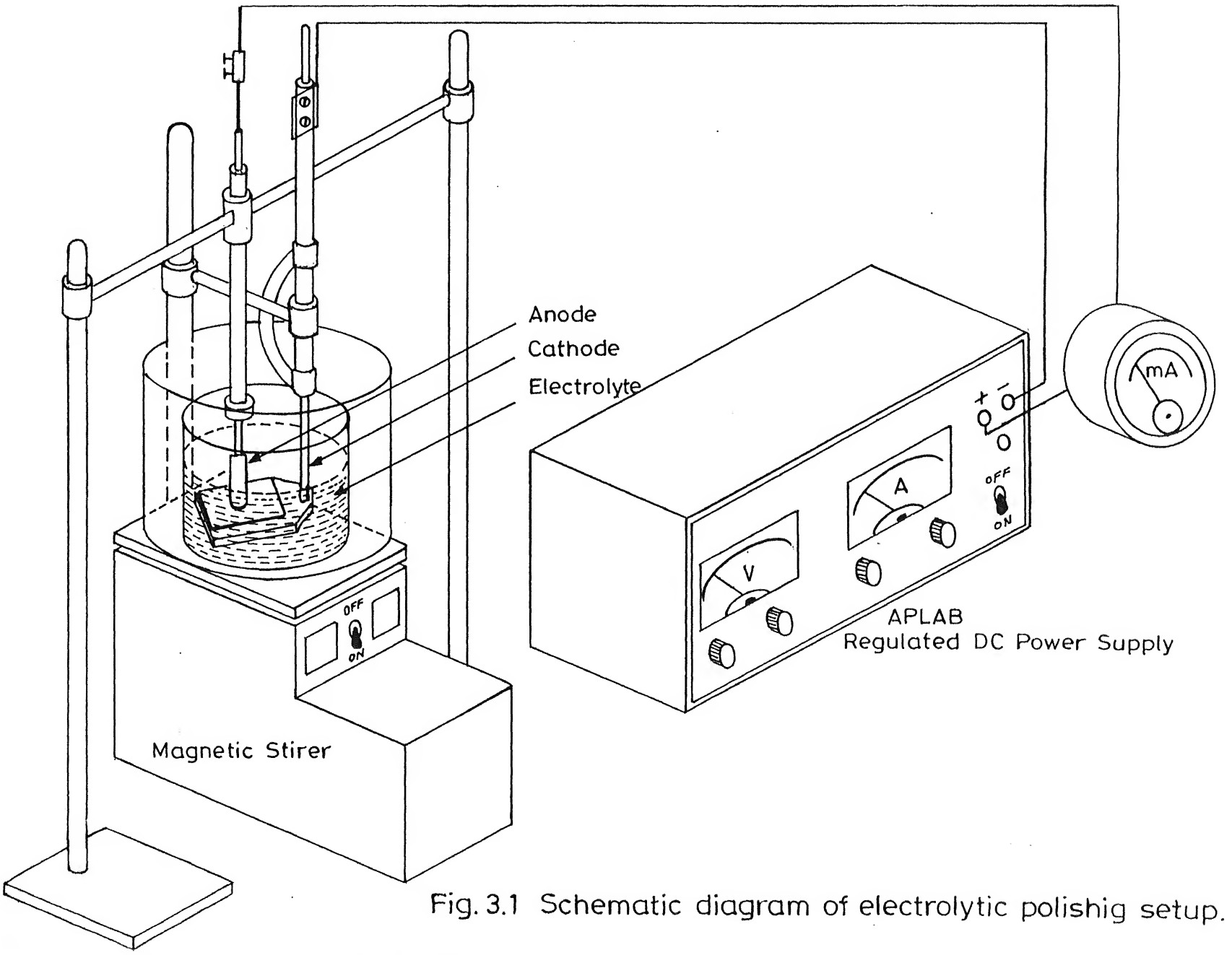


Fig. 3.1 Schematic diagram of electrolytic polishig setup.

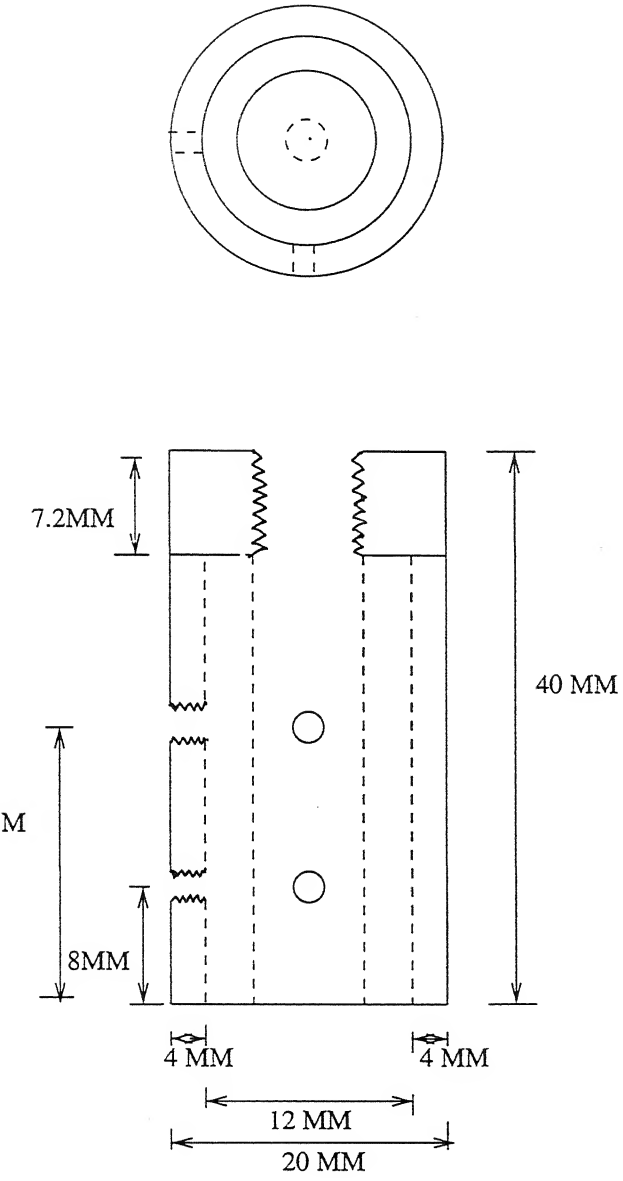


Fig No. 3.2 Teflon Sample holder

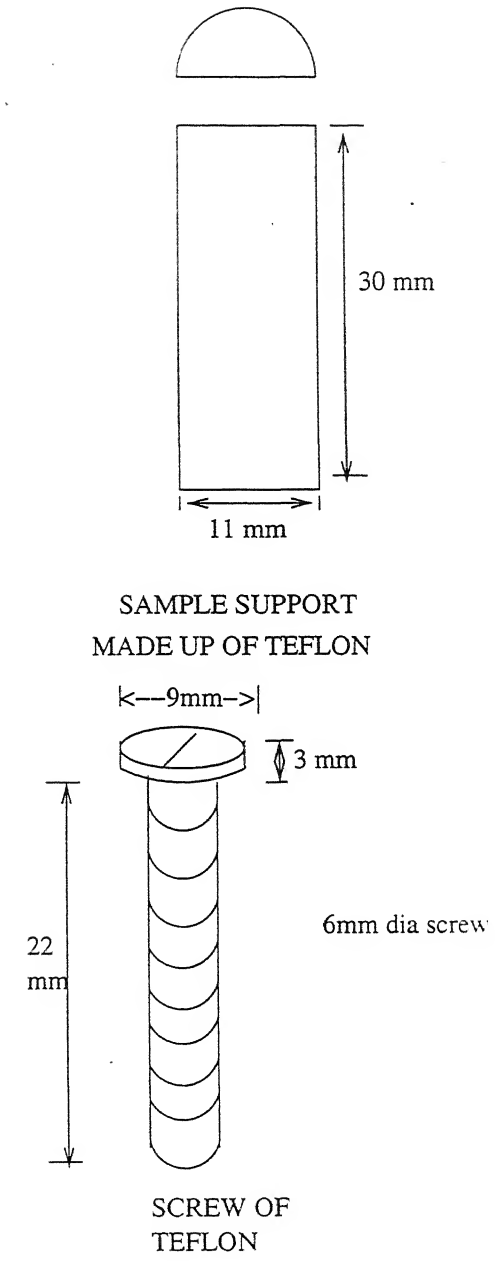


Fig No. 3.3 Teflon Screw and Teflon Supprt

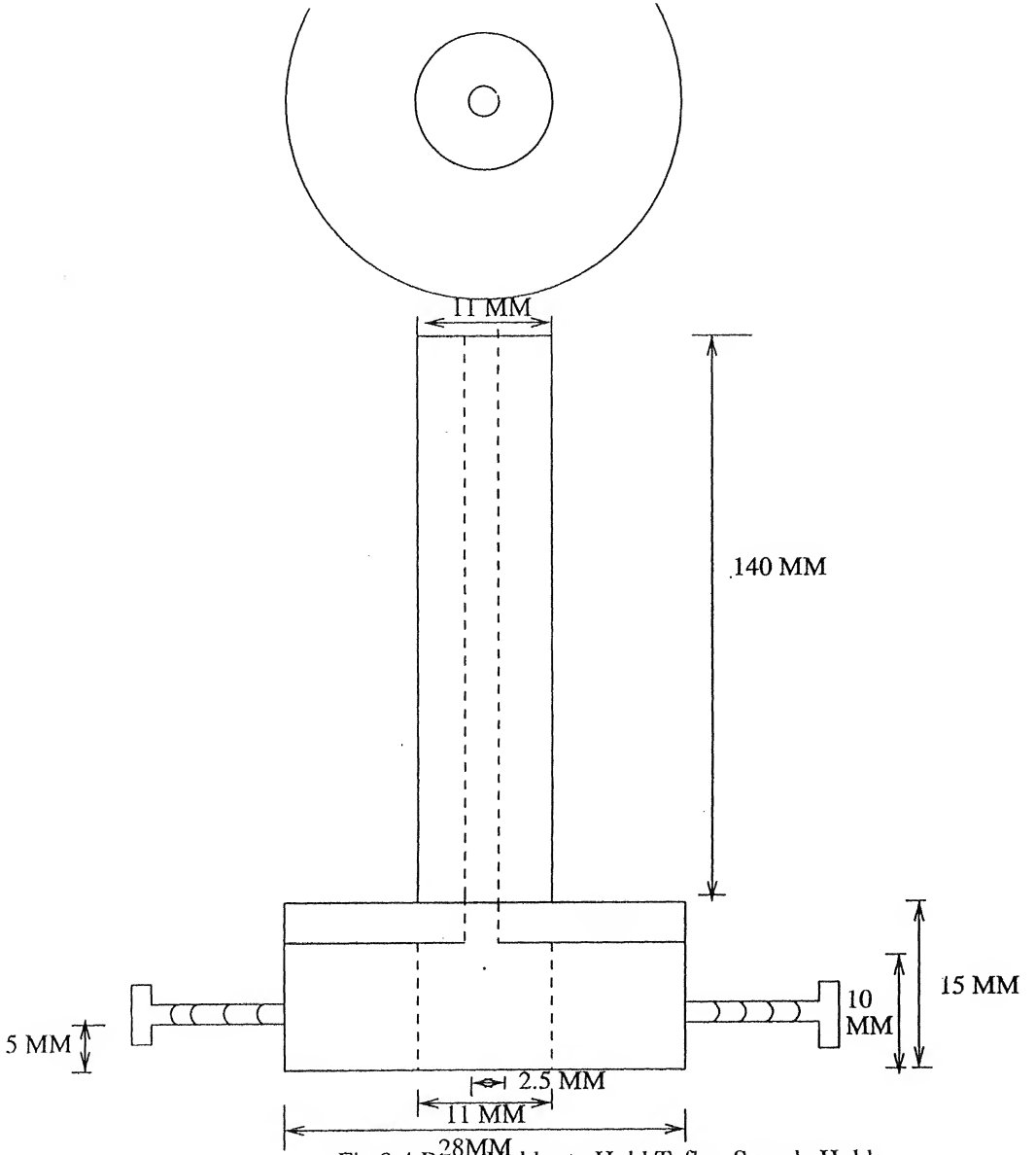


Fig 3.4 Brass Holder to Hold Teflon Sample Holder

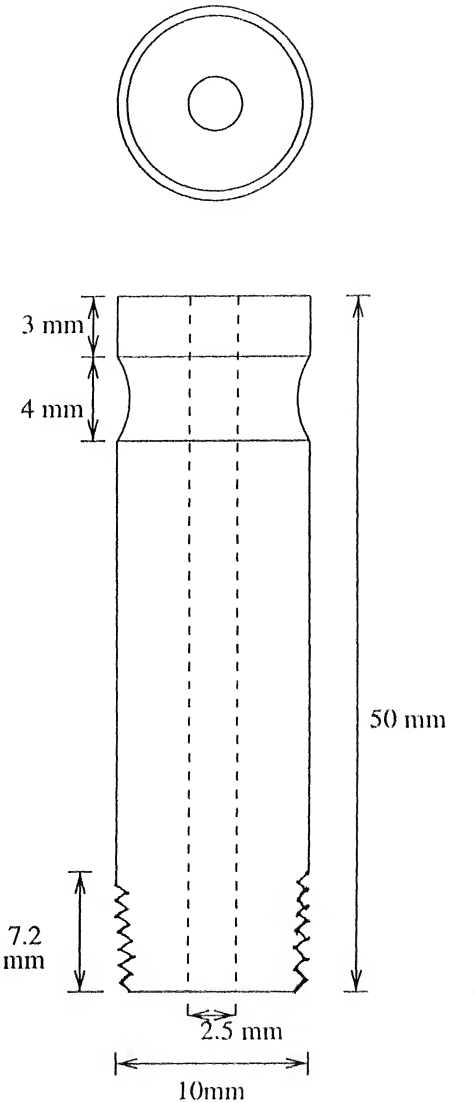


Fig. No. 3.4 Threaded teflon rod to hold sample holder

3.2.2 Electrical Connections: For electrical connections the cathode i.e. tin rod was connected to negative terminal the cc – cv source. For connecting the anode to positive terminal a copper wire was passed through the brass tube and Teflon holder. This wire goes into the sample holder where it is brazed to a piece of silver sheet. The silver sheet touches the samples and lies between the sample and the sample supporting Teflon blank. The top end of copper wire is connected to positive terminal of the voltmeter. The electrical connections are shown in figure 3.1.

A magnetic stirrer was used for stirring the electrolyte during electrolytic polishing.

3.2.1.2 Electrolyte: The electrolyte [20] used for polishing was:

Electrolyte	Cell Voltage	Time
70% ethanol (absolute) 12% distilled water 10% 2-butoxyethanol, 8% perchloric acid(60%)	30 – 65 volt	15 – 60 sec

The working condition for electrolytic polishing was established after many trial and is given below

Cell Voltage	Current	Time
40 volt	150 mA	45 sec

After electropolishing, individual grains and the dendritic structure were visible without etching. One of the larger grains in each sample was located and was exposed to X-ray. The location of this grain in the transverse section noted in the

logbook and a spot in this grain was selected; that very spot was exposed to x-ray with eyeball judgement.

3.3 OBTAINING X-RAY PATTERN

For obtaining Laue back reflection pattern, samples were exposed to X-rays of white radiation. The X-ray film was kept in a cassette of dimension 8.3 X 10.1 cm. The X-ray film was cut to this dimension as far as possible. The X-ray film was kept in a black envelope to protect it from visible light. A small cut was made at the top right corner on each of the film for the identification and for future analysis. The samples were exposed to X-ray of Cu K_α radiation and at 20 mA and 30 kV. The voltage of the X-ray tube could not be increased due to some technical limitations of the system. The samples were exposed for about 10 hours. It was found that at this voltage the patterns were not very sharp and the spots were week. For one of the samples exposure time was increased to 14 hours and which resulted in better pattern.

The X-ray film was developed in a dark room and dried for further work.

3.4 OBTAING THE INPUT DATA

The input data required for the program are obtained from the Laue back reflection pattern. The input data, which are obtained from the pattern, are

1) x and y coordinates of each of the selected spots
2) No. of zones and zone angles

3.4.1 Obtaining X and Y Coordinates of Spots

For obtaining x and y coordinates of spots, the X-ray pattern is kept on a glass plate and a tracing paper fixed over the film. By illuminating the glass plate

from below the position of each spot becomes clear and traces of spots are made on the tracing paper. After transferring spots on tracing paper the x and y-axis are also drawn on the tracing paper. The origin is plotted at the center of the hole in the film. Thus a replica of the film is prepared.

This replica is then superimposed over a graph paper with 1.0 mm division. Then the x and y coordinates of each spot is measured with $\pm$1 mm accuracy.

3.4.2 Obtaining Number of Zones and Zone Angles

For obtaining number of zones and mutual angles between zones, stereographic projections were made for each of the sample. For drawing stereographic projection the Greninger angle, γ and δ, for each spot is measured with the help of Greninger chart for specimen film distance 3 cm. The spots belonging to one great circle are located and all possible great circles are traced, with each great circle containing minimum of three spots. The spots belonging to one great circle are known as spots of one zone. The great circle containing more than three spots are considered as low indexed zones. A minimum of two zones has to be located. The spot lying at the intersection of two or more than two zones are considered as the prominent spots and are of low indices.

After selecting few important zones the zone axes for all the zones are located on the stereographic projection. After locating zone axes the mutual angle between the zone axes are measured on the stereographic projection.

The proper selection of the desired number of spots is important for the indexing of the spots. The spots are numbered arbitrarily in the beginning and the stereographic projection for the sample is made. After making stereographic projection the spots are renumbered. The spots lying at the intersection of two or more than two zones are renumbered first and than out of the remaining spots the ones closer to the center and/or darker are numbered in that sequence. A minimum five or six spot are selected so that they involve atleast two or three zones.

CHAPTER 4

COMPUTER PROGRAM

4.1 TECHNIQUE ADOPTED:

This is an iterative scheme of matching the interplanar angles between the experimental spots of the Laue back reflection pattern and theoretical angles. The experimental angles are calculated using the x and y coordinates of spots on the Laue back reflection pattern and theoretical angles are calculated using the crystallographic expression for body centered tetragonal system after assigning Miller indices. The scheme assigns indices to spots such that the difference between experimental and theoretical angles remains within three degrees. At the same time the program also checks the zone condition, that is spots belonging to one zone should satisfy the zone condition. The scheme determines the Miller indices of the zone axes for the zones. After assigning indices to zone axes the zone angle between the zone axes are calculated using the crystallographic expression for body centered tetragonal system. The difference between experimental (measured on the stereographic projection of the LBR pattern) and theoretical zone angles are calculated. If this difference is found to be within the allowed error limit then the program assigns indices to spots. The program assigns indices to spots only after both the conditions are satisfied.

4.2. MEASUREMENT OF MUTUAL ANGLES

4.2.1. Measurement of Experimental Angle

The experimental angles refer to the mutual angles between the spots obtained from their x and y coordinates. The method adopted is one developed by Christensen [7, 8]. The computation of angles associated with pairs of

reflections is accomplished in a perfectly straightforward manner. The geometry involved in the calculation of the angles is explained in figure (4.1). The film plane is assumed to have the X and Y-axes. The beam direction is taken as the Z-axis. The specimen to film distance is taken as D. Each spot on the film is assigned x and y values with the origin at the point where the X-ray beam intersect the film. The angle $2\phi_{ij}$ is the angle between two reflections i and j from the sample S. These two reflections have x and y values as (x_i, y_i) and (x_j, y_j), with respect to the origin O. From figure 4.1 a,

$$L_1^2 = Xi^2 + Yi^2 + D^2 \quad (1)$$

$$L_2^2 = Xj^2 + Yj^2 + D^2 \quad (2)$$

$$L_3^2 = (Xi - Xj)^2 + (Yi - Yj)^2 \quad (3)$$

Figure 4.1 b gives another view of the two spots and the sample, in which

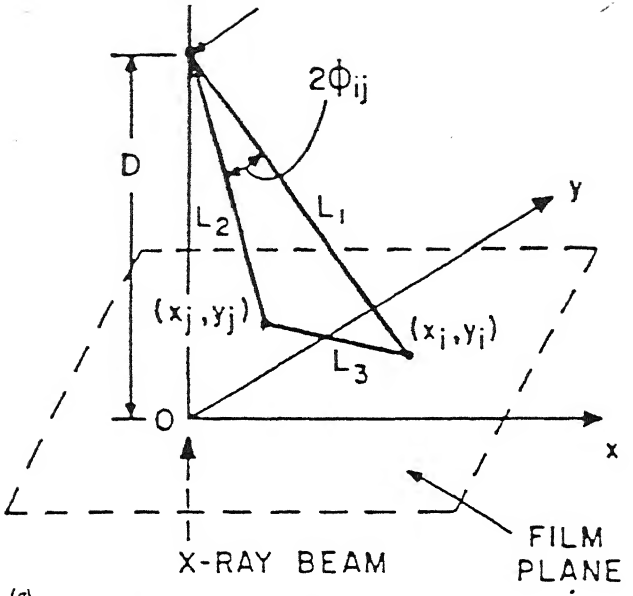
$$L_4 = \frac{L_1^2 - L_2^2 + L_3^2}{2L_3} \quad (4)$$

$$L_6^2 = L_1^2 - L_4^2 \quad (5)$$

$$2\phi_{ij} = \tan^{-1}(L4/L1) + \tan^{-1}((L3-L4)/L6) \quad (6)$$

$$\phi_{ij} = \tfrac{1}{2} (\tan^{-1}(L4/L1) + \tan^{-1}((L3-L4)/L6)) \quad (7)$$

ϕ_{ij} represents the angles between the spots, i and j. These angles are measured for all spots to be indexed. These are the experimental angles ϕ_{ij} , i=1, 2, .., N-1 and j = i +1 ,i+2, ..., N, where N is the total number of spots to be indexed.

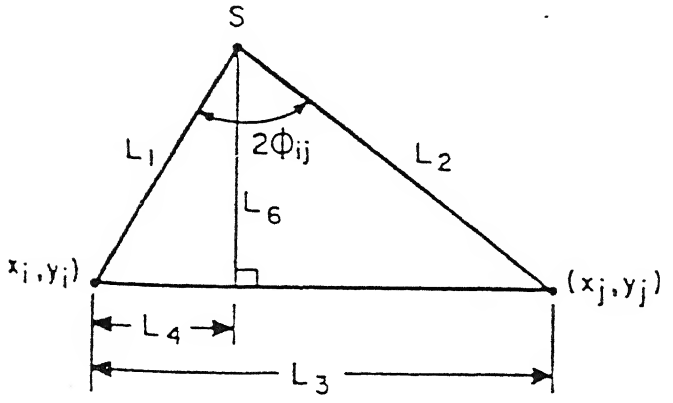


(a)

$$L_1^2 = x_i^2 + y_i^2 + D^2$$

$$L_2^2 = x_j^2 + y_j^2 + D^2$$

$$L_3^2 = (x_i - x_j)^2 + (y_i - y_j)^2$$



(b)

$$L_4 = \frac{L_1^2 - L_2^2 + L_3^2}{2L_3}$$

$$L_6^2 = L_1^2 - L_4^2$$

Fig 4.1 Measurement of experimental angle using coordinates of spots

4.2.2 Calculation of Theoretical Angles

For a set of indices assigned to two spots, the theoretical angle between them is calculated using the crystallographic expression for the body centered tetragonal system, since tin has a body centered tetragonal structure at room temperature. These angles are designated as ϕ_T. the expression for calculating the theoretical angle is given below

$$\cos(\phi_{ij}) = \frac{(h_i h_j + k_i k_j)(c/a)^2 + (l_i l_j)}{\sqrt{[(h_i^2 + k_i^2)(c/a)^2 + l_i^2][(h_j^2 + k_j^2)(c/a)^2 + l_j^2]}} \qquad (8)$$

where (h, k, l) are the assigned indices to spots i and j.

4.2.3 Calculation of Zone Angles

The scheme also calculates zone angles, after assigning indices to zones, between the assigned zone axes. The formula for the calculation for angles between zone axes is given below

$$\cos(\phi_{ij}) = \frac{(u_i u_j + v_i v_j)/(c/a)^2 + (w_i w_j)}{\sqrt{[(u_i^2 + v_i^2)/(c/a)^2 + w_i^2][(u_j^2 + v_j^2)/(c/a)^2 + w_j^2]}} \qquad (9)$$

Where (u, v, w) are the assigned zone axes.

Theoretical zone angles and experimental zone angles, calculated after indexing of spots, are checked and if their difference lies within error limit then indexing of spots proceeds.

4.3 SELECTIONS OF INDICES

The index search for this program is restricted in between (6 6 6) to ($\bar{6}\ \bar{6}\ \bar{6}$), since a judicious choice of spots on the film will normally ensure that their indices lie within this range. The total number of indices possible in this range is about 2200. For a body centered crystal, from the structure factor expression, planes with (h + k + l) = odd do not produce any reflections.

The structure factor expression for body centered tetragonal system is,

$$F = \sum_{i=1}^{n} f_i \, e^{-2\pi I(hu + kv + lw)}$$

where f is the atomic scattering factor, h, k, l are the Miller indices of the planes and u, v, w are the positions in the unit cell of the crystal. For a body centered crystal the atom positions are (0 0 0) and (1/2, 1/2, 1/2). Hence the structure factor expression for a body centered crystal is,

$$F = f\left(1 + e^{-\pi i(h + k + l)}\right)$$

For (h + k + l) = even, $F = 2f$, $I \alpha F^2 = 4F^2$

For (h + k + l) = odd, $F = 0$ and $I = 0$.

Hence all those indices with (h + k + l) = odd are excluded from the range of indices. This exclusion results in a total of 1099 possible set of indices in this range.

Considerable savings in search time can be achieved without any loss in generality if scalar multiple of previously considered indices are excluded from the search. In general if (h k l) is considered then (nh, nk, nl) need not to be considered where value of n varies from -6 to –2 and 2 to 6. These exclusion leads to possible set of indices in this range to 868. So if (110) is considered then all the scalar multiple of this such as (2 2 0), (3 3 0), (4 4 0), (5 5 0), (6 6 0) and similarly all negative multiple such as ($\bar{2}\ \bar{2}$ 0), ($\bar{3}\ \bar{3}$ 0) etc. are neglected.

The set of selected indices within this specified limit is arranged from lower indices to higher indices, so that the lower indices are assigned first to the spots. For a given index set, (h k l), larger the interplanar spacing, lower will be the indices. Based on this criteria, the index sets are arranged in descending order of d spacing, which is calculated for each index set from the expression,

$$\frac{1}{d^2} = \frac{h^2 + k^2}{a^2} + \frac{l^2}{c^2}$$

Where h, k, l are the indices and a and c are the lattice constants.

4.4 CRITERIA FOR FIXING THE ERROR LIMIT:

The computer program is developed with an allowable error limit of ± 3 degrees, in matching the interplanar angles. The error limit set is based on the error likely to have been introduced during the measurement of input data. These are as follows:

(i) The spots on the X-ray film generally do not have an exact location. The identification of the exact location is difficult, if the spots have fuzziness.

(ii) The experimental angles are calculated with the standard value of D (film to sample distance) as 3.0 cm. But, while fixing the sample for X-ray exposure, the distance of separation between the sample and the X-ray film could be any where between 2.8 to 3.2 cm. With enough care it will be possible to fix the distance within this range. Hence this limit is kept in optimizing the distance of separation. But since, the experimental angles are calculated initially with D as 3.0 cm, there will a certain amount of error introduced in the experimental angles.

(iii) The X-ray beam may not be passing through the center of the hole punched in the film and hence the x and y axes for the spots on the film may not be parallel to the edges of the film and the center of the hole may not coincide with the true origin for the spots.

These errors can vary the measured experimental angles. Considering all these factors, the error limit is kept at 3.0 degrees.

4.5 SCHEME OF INDEXING:

The scheme finds out trial indices for N spots, such that all the experimental and theoretical angles are within an allowable error range. The error criterion is defined as,

$$|\phi T - \phi E| \leq \text{ERROR} \qquad (11)$$

The allowable error range is fixed as ± 3.0 degrees. All the spots are initialized with the indices ($\bar{1}\ \bar{1}\ 0$), which is the first indices set to be assigned. The scheme tests the angular match between a pair of spots at any stage. First it checks the angular match between spots number 1 and 2. If the angle between them is not matching with the experimental angle then indices of spot 2 is changed and next indices from the possible set is assigned to the spot 2 and angle between them is again checked. If with all the possible set of indices, suitable match is not found then indices of spot 1 is changed and it is assigned next to previous assigned indices to spot 1 from the possible set. If match is found, then keeping indices of 1 and 2 fixed spot 3 is assigned with the indices and angular match between spot 3 and 1 is checked. If angular match between them is not found then indices of spot 3 is changed to next indices from the possible set of indices. If all the indices of the set are checked and no suitable

match is found then indices of spot 1 is changed and the spot 1 is assigned next to previous assigned set of indices. Assigning of new indices to spot number 1 essentially means that search of indices for spot starts from the beginning. That is, the indices for spot number 2 are searched and then the process continues as mentioned above. If angular match between spot 3 and 1 is found then the scheme checks the angular match between spot 2 and 3. If angular match between spot number 2 and 3 is not within the error limit then indices of spot number 3 is changed by assigning the next set of indices is assigned to it and angular match between spot 3 and 1 is again checked. If angular match between spot 2 and 3 is not within the error limit, although angular match between spot 3 and 1 is within error limit, the indices of spot number 3 will not be excepted. This way indexing of spots carries on. Assigning indices to any spot essentially mean that it should satisfy the angular match with all the previous spots. That is, if spot number 5 is being assigned indices then it should satisfy the angular match with spot 1, spot 2, spot 3, spot 4.

The scheme also checks the zone condition. If two spots belong to one zone then it should satisfy the zone condition. For this, zone checking is carried out in the program. If spots 1, 2 and 4 belong to one zone then program calculates their determinant using their indices. If at any time spot number 4 is being assigned indices then the determinant of the spots number 1, 2 and 4 is checked. If the determinant value does not come to zero then the indices of spot number 4 will not be excepted and spot number 4 will be assigned next to previous assigned set of indices. If determinant value come to zero then the scheme calculates the zone angle between zones. If difference between zone angles, calculated by program after assigning indices termed as theoretical zone angle, and measured from stereographic projection, termed as experimental zone angle, lies within error limit then the scheme assigns indices to spots and gives their zone axes and angle between these zone axes. If the difference between zone axes do not lie within error limit then the indices of the last spot lying on the zone is changed and searching starts from there.

4.6 DETERMINATION OF CRYSTALLOGRAPHIC ORIENTATION

The scheme, after indexing the required number of spots gives the indices of zone axes also. From these indices of zone axes, the orientation can be determined easily. The samples used in the present work were Sn-Bi alloys, which have their primary dendrites roughly parallel to the growth direction, which has been reported as [1 1 0], [1 $\bar{1}$ 0] and [001] in the literature [1, 2, 3]. The angle between the indexed zone axes and [1 1 0] is calculated. The method of determining the orientation is as follows: the stereographic projection of the pattern is fixed on a wulff net, such that the X and Y axes of the projection coincide with the E-W and N-S directions of the net respectively. The pole of one of the axis is first rotated with respect to the N-S axis to bring the pole on the circumference of the net. The angle by which the pole is rotated about the N-S axis is noted. The pole is then made to coincide on either the North or South Pole of the net. With this position of the pole, the small circle, which is at the required angular distance from the pole, is traced. Few points are marked on this small circle. Now the pole is rotated back with respect to the axis perpendicular to the projection. The pole is again brought to its initial position by rotating with respect to the E-W axis by the same number of degrees, by which it was rotated before. The points, which are marked on the small circle, are also rotated by the same number of degrees about the N-S axis. The arc containing the new position of these points is traced, which in turn will be at the required number of degrees from the zone axis. This process is carried out for the other zone axis also. The intersection of the two small circles traced for each pole, fixes the location of the desired zone axis ([1 1 0]). Now the angle between this axis and the center is measured by aligning the two poles to lie along the equator. In terms of this angle, the orientation of the crystal is

expressed. The methods for determination of orientation is given in figure number 4.2 in the next page.

4.7 STRUCTURE OF PROGRAM

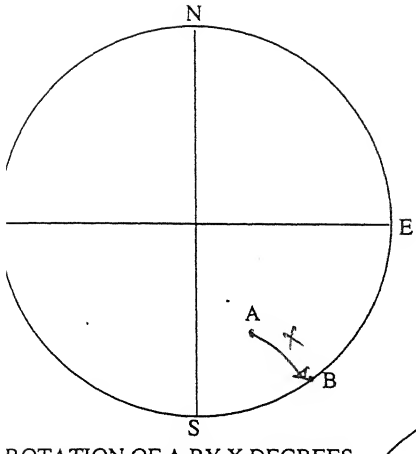
The computer program developed in 'C' language is divided into a number of functions, with each function doing a specific task. The functions are arranged in a sequence in which the tasks are to be performed. The functions are called from main program as well as from a function. This section contains basic outline of the program and the flow charts of these functions are given in appendix A.

4.7.1 Input Data

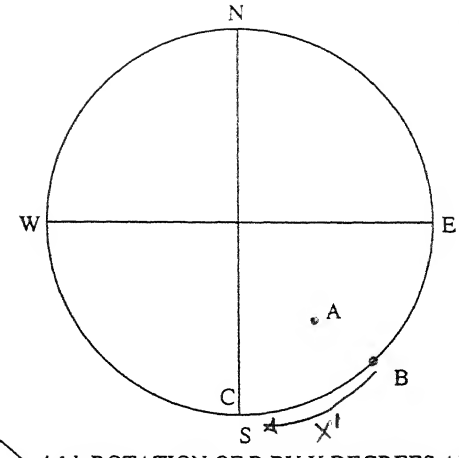
The input data required be listed below in the same order in which they are fed into program

(1) The sample number (S)
(2) Total number of spots to be indexed (N)
(3) The coordinates of N spots
(4) Number of zones to be indexed
(5) Spots belonging to one zone
(6) Experimental zone angle between zones (ZEXP) for all the zones

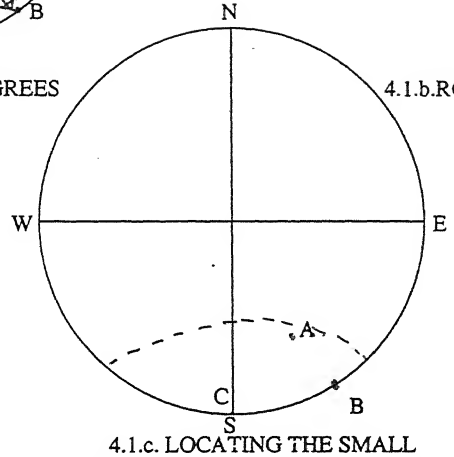
Additional input data required are, error limit allowed, the maximum and minimum limits of Miller indices to be assigned to the spots (MAX, MIN), c/a ratio of the crystal (d), the separation between sample and X-ray film (D). These data are defined globally and can be availed by any function at any stage of program.



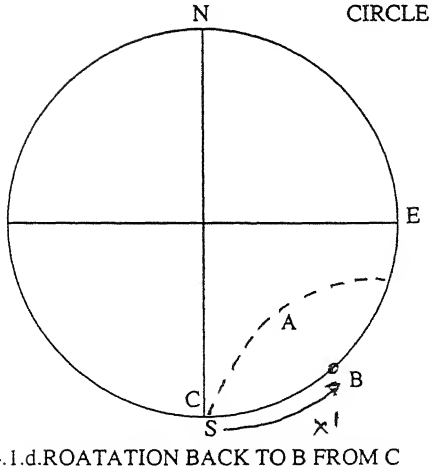
ROTATION OF A BY X DEGREES BOUT N – S AXIS, TO B



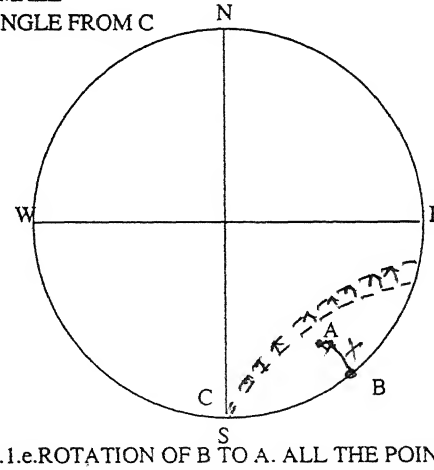
4.1.b.ROTATION OF B BY X DEGREES ABOUT THE PERPENDICULAR AXIS, TO C



4.1.c. LOCATING THE SMALL CIRCLE AT THE DESIRED ANGLE FROM C



.1.d.ROATATION BACK TO B FROM C



4.1.e.ROTATION OF B TO A. ALL THE POINTS ARE ROTATED ABOUT N–S AXIS

Fig 4.1: Determination of growth direction

A maximum of 10 spots can be indexed. This program can be easily modified for more than 10 spots. The x and y coordinates of each spot to be indexed should be obtained with an accuracy of ± 1mm. The stereogram for each sample is to be prepared and spot belonging to one zone is identified. Angle between zones is measured from this stereogram.

Following sections consist a short detail of each function and the flow chart of each function is attached in APPENDIX A.

4.7.2 Function EXPANG

This function calculates experimental angles between spots using their coordinates. The calculated angles are stored in expang[i][j] permanently which are defined globally, that is these value can be used anywhere in the program. This function is called only once from the main program and it returns back after calculating all mutual angles between spots. The input data for this function are the x and y coordinates of spots which are made available to it through the argument. Other data required for this function are the separation between X-ray film and sample, and the value of pi. These values are defined globally so these are obtained directly.

The geometry of calculation of angles is given in section 4.1.1. This function takes the value of coordinates from main program and stores it in an array. Then it calculates the mutual angles between spots and stores it in an array expang[i][j], where i and j represents the spot number.

4.7.3 Function TANGLE

This function calculates theoretical angle between spots using crystallographic expression for body centered tetragonal system. The

crystallographic expression for calculation of angle for body centered tetragonal system is given in section 4.1.1. The input data for this function are two sets of Miller indices at any time and the c/a ratio is made available to this function from main program. This function is called from main program many a time during searching of Miller indices for spots. The calculated angle is returned as angle, which is stored in tangle[i][j], in the main program representing theoretical trial angle between spots i and j.

4.7.4 Function INDICES

This function generates all the possible set of Miller indices for a body centered tetragonal system within a specified maximum and minimum limit of Miller indices. In the present work we restricted our indices search in the range of (666) and ($\bar{6}\ \bar{6}\ \bar{6}$). The entire possible Miller indices set in this range is generated in this function and is arranged in descending order of their interplanar spacing. This function is called only once from the main program and it returns to the main program and all the Miller indices are stored in the array of h[], k[], l[] globally.

The selection of Miller indices is a straightforward technique. Three counters i1, k1, l1 are used for this. This function generates only those indices for which the sum of their h, k, l are even. That is all the Miller indices set whose sum of h, k, l is odd are neglected. The counter l1 is varied from minimum limit of Miller indices, -6 for our program, to the maximum limit keeping other two counter fixed. Once the counter l1 changed from –6 to +6 the counter k1 is increased by step one. The initial value of i1, k1, l1 is kept at minimum limit of Miller indices i.e. –6. So, if the entire l1 is changed from –6 to +6 then the value of counter k1 is increased. Thus for any one value of i1, k1, all the possible set of Miller indices are generated. Similarly other counters are also changed so that all the possible set of Miller indices within this range is generated and are stored in the array h, k, l. After selection of indices these

indices are arranged in descending order of their interplanar spacing. The planes with lower indices have larger interplanar spacing. The crystallographic expression for the calculation of interplanar spacing is given in the section 4.2.2.After arranging the Miller indices according to their interplanar spacing this function calls another function REMOVE () which removes all the set of indices which are scalar multiple of previous stored indices.

4.7.5 Function REMOVE

This function is called only from the function indices and it removes all indices, which are scalar multiple of previous indices. The number of times this function is called is depends on the possible number of sets of Miller indices in the range used in the program after considering structure factor condition. The input data for this function is the Miller indices at any time and this function checks whether this Miller indices is scalar multiple have any previously stored Miller indices. Suppose at any time Miller indices (2 2 2) is under check then this function will check whether this set of indices is scalar multiple of any previous Miller indices set and, in this case, if it found that this set is not a scalar multiple of any previous Miller indices set then this function return a value which instructs the calling function to store this set in the selection list of Miller indices. Suppose at any time Miller indices set (4 4 0) is under check and since this set is scalar multiple of Miller indices set (1 1 0) so this function will return a value which will instruct the calling function that this set of Miller indices is scalar multiple of already stored Miller indices set so this set will not be included in the search list of Miller indices. This function also removes the Miller indices set (0 0 0) from the list of search. So at any time this function takes a single set of Miller indices and checks the condition of scalar multiplicity and according to the result it returns the value to the calling function to store or reject the Miller indices under consideration.

4.7.6 Function ZONE

This function is called from the function check described in section 4.7.7 for zone checking. This function takes the assign indices set as the input data and checks the zone condition. If three spots belong to one zone then taking their h, k, l as the individual members their determinant is calculated. If the value of determinant comes out to be zero then the zone condition is satisfied. This function returns different value depending on the condition. If their determinant comes out to be zero then it returns a value 1 which indicates that the zone condition is satisfied otherwise the function returns 0 and the program continue next search for the spots. This function is called many times from the function check till all the conditions are satisfied.

4.7.7 Function CHECK

This function is called from the main program and this checks all the conditions required for indexing. The conditions, which are to be checked, are the interplanar angles and zone condition. The input data for this function at any time is the assigned Miller indices set to any spot and the spot number, which is being assigned. The detailed scheme of indexing of patterns has been discussed in the section 4.5. This function then checks the interplanar angles between spots, which are already assigned with Miller indices. The scheme matches the theoretical and calculated interplanar angles between spots and if the error condition is satisfied then zone condition is also checked. If at any time spot number 5 is being assigned Miller indices then the input data for this function is 5 and the Miller indices of the spots which have been assigned. Then this function matches the interpalnar angles between spot 5 and 1, spot 5 and 2, spot 5 and 3, and spot 5 and 4. If the difference between their theoretical and experimental angles remain within error limit then this function returns a value to calling function to continue with assigning to next spot i.e. 6. If the

difference in interplanar angles is not within error limit then this function will return a value which will instructs the calling function to assign new indices to the spot number 5. If all the possible set of indices is assigned to spot 5 and no suitable match is found then the indices of spot with which the interplanar angle condition is not satisfied is changed and the search for the indices continues. The function also checks the zone condition. If spot 1, 2, and 3 belong to one zone then at the time of assigning indices to spot number 3 the zone checking is also carried out in this function. The function zone is called from this function and the argument for the function zone is the Miller indices of spot 1, 2 and 3. If their determinant is zero then the function zone will return a value which will instruct the program that the zone condition is satisfied and the scheme will continue with indexing of next spot. If the zone condition is not satisfied then the function zone will return a value which will instruct the calling function that the zone condition is not satisfied and so the indices for the spot number 3 is not acceptable. Then the function check will return a value to calling function, which will instruct the function to assign new indices to spot number 3. If with all the possible set of Miller indices set the zone condition is not satisfied then the spot just previous to it that is spot number 2 in present case is changed and fresh search of Miller indices for the spot number 3 is done. So assigning of Miller indices to any spot means that it should satisfy the interplanar and zone condition. When the last spot belonging to last zone is assigned and zone condition for all the zone is satisfied then this function assign indices to the zone axes for each zone and also calculates zone angles between zones. The indices zone axes is determined using the expression given below

$$ZH[i] = (k_i * l_j - l_i * k_j)$$

$$ZK[i] = (l_i * h_j - h_i * l_j)$$

$$ZL[i] = (h_i * k_j - k_i * h_j)$$

After calculating zone angles between zones, which are termed as theoretical zone angles, the difference between theoretical and experimental zone angles is calculated. If the difference between theoretical and experimental zone angles remains within error limit then this function returns a value which will instruct the calling function (main program in this case) to call function print which will print all the results in formatted manner. If the difference in zone angles is not within the error limit then this function will return a value to the calling function which will assign new indices to the last spot and the fresh checking will be carried out.

4.7.8 Function PRINT

This is the last function called from the main program after all the conditions are satisfied. This function prints results in a formatted manner. This function gives all the required results in a well-structured form. This function gives the result in a sequence, which is given below.

1. Sample number.
2. Number of spots to be indexed.
3. Range in which Miller indices are searched.
4. Coordinates of spots.
5. Indices of spots.
6. The experimental angle and theoretical angles between spots and difference between them.
7. Spots belonging to one zone and their zone axes.

Experimental and theoretical zone angle between zones and difference between experimental and theoretical zone angle

4.8 MAIN PROGRAM

The main programs first reads all the input data and then calls function in a sequence to which work has to be done. The input data for this program are of two types.

a) Some of the input data, which remain constant for the entire sample, are stored permanently. Some of the data are defined globally so that it can be used from any function and from anywhere in the program.

b) Other type of input data is different for different sample. The sequences in which the input data have to be fed are given below.

1. Sample number.
2. Number of spots to be indexed (N).
3. Coordinates of N spots.
4. Number of zones to be considered.
5. Spots belonging to one zone.
6. Experimental zone angle between zones.

These data can be fed either through keyboard or through input file. The sequence has to be maintained during inputting the data.

The main program first calls function INDICES ( ) which generates all the possible set of Miller indices and stored globally. Then the main program calls function EXPANG( ) which calculates mutual angles using the coordinates of spots and angles are stored in variable expang[i][j] where i and j are the spot number. These experimental angles are stored permanently, which are defined globally for its use in any function.

The program then starts searching Miller indices for spots. The first Miller indices assigned to any spot is [$\bar{1}$ $\bar{1}$ 0], because this is the first Miller indices set which is stored in the possible list of indices after considering all the conditions including interplanar spacing. After assigning indices to spot number1, the program assigns indices to spot 2. Spot no. 2 is also first assigned with indices [$\bar{1}$ $\bar{1}$ 0]. Then the theoretical angle between spot 1 and 2 is

calculated by calling function TANGLE( ) which returns the theoretical angle between spot 1 and 2 and this angle is stored in tangle[i][j] where i and j are spots number. After that the difference between theoretical and experimental angle is measured. If the absolute value of this difference between these two angles comes out to lie within error limit then the program will assign indices to spot 1 and 2 and search for next spot will be carried out. If the angle difference is out of error limit then the scheme will assign next set of Miller indices to spot number 2 keeping the Miller indices of spot 1 fixed and the theoretical angle between these two spots is calculated and their difference is measured. So, the scheme will search the Miller indices for spot two till the angular match is found. If all the possible set of Miller set is assigned but no suitable match is found then indices of spot 1 will be changed and spot 1 will be assigned with next set of indices from the list. If the match is found then keeping the Miller indices of spot 1 and 2 fixed scheme starts searching Miller indices for spot number 3. The spot number 3 also will be assigned first indices as $[\bar{1}\ \bar{1}\ 0]$ and the angular match between spot 1 and 3 is carried out as explained above. If the angular match between spot 3 and 1 is found then the main program calls function CHECK (t2, H, K, L) for the checking. Here the argument t2 is the number of spot which is under assignment and H, K, L are the arrays of Miller indices of spots which have already been assigned with indices. In present case the value of t2 is 3 and the array H, K, L contains the Miller indices of spot 1, 2 and 3. So, if at any time spot number 6 is being is assigned then t2 will be 6 and H, K, L will contains all the Miller indices set of spot from 1 to 6. The function CHECK( ) checks the interplanar angle and zone condition for the spots. If the interplanar and zone condition is satisfied then the function CHECK ( ) will return a value which instructs the main program to continue search for the next spot. If any condition is not satisfied then the scheme will return a value which will instructs the program to assign new Miller indices to spot number which is under checking. If for all the possible Miller indices is assigned to spot which is under checking and no

match is found then the indices of spot just prior to this spot is changed and assigned new indices and angular check between spot 1 and the last spot whose indices have been changed is checked out and the process of checking is carried out as explain above.

If all the condition is satisfied, i.e. interplanar angles and zone condition is satisfied then the function CHECK ( ) will return a value which instructs the program to call function PRINT ( ) which gives results including input data in a formatted manner.

CHAPTER 5

RESULTS AND DISCUSSION

A computer code in 'C' language was developed and the developed code was used for indexing of two Laue pattern from the body centered tetragonal Sn- Bi alloys. The composition of alloy for both the samples was Sn-2 wt% Bi.

The Laue back reflection patterns of the two samples with their x and y coordinates of spot are given in appendix B. Both patterns were indexed with the help of computer program and the output of the program for both patterns are given in appendix C. The stereographic projection of both the samples are icluded in appendix D.

The indices assigned by the program are given below. The indices for the sample number 33 and the zone axes are:

INDICES OF SPOT [1] IS →	1	1	2
INDICES OF SPOT [2] IS →	4	4	6
INDICES OF SPOT [3] IS →	3	3	4
INDICES OF SPOT [4] IS →	1	0	3
INDICES OF SPOT [5] IS →	1	0	1
INDICES OF SPOT [6] IS →	4	3	5

Spot 1, 2 and 3 belong to zone $[\bar{1}\ 1\ 0]$

Spot 3, 5 and 6 belong to zone $[3\ 1\ \bar{3}]$

ZONES	ZONE ANGLE		
	THEORETICAL	EXPERIMENTAL	DIFFERENCE
$[\bar{1}\ 1\ 0]$ and $[3\ 1\ \bar{3}]$	113.4	112.0	1.4

The indices of the spots and the zone axes for the sample number 35 are:

INDICES OF SPOT [1] IS → $\bar{4}\ \bar{5}\ 1$

INDICES OF SPOT [2] IS → $\bar{5}\ \bar{6}\ 1$

INDICES OF SPOT [3] IS → $\bar{2}\ \bar{5}\ 1$

INDICES OF SPOT [4] IS → $\bar{1}\ \bar{3}\ 2$

INDICES OF SPOT [5] IS → $\bar{3}\ \bar{5}\ 4$

INDICES OF SPOT [6] IS → $\bar{2}\ \bar{3}\ 3$

Spot 1, 2 and 4 belongs to zone $[\ 1\ \bar{1}\ \bar{1}]$

Spot 3, 5 and 6 belongs to zone $[\ \bar{3}\ 1\ \bar{1}]$

ZONES	ZONE ANGLE		
	THEORETICAL	EXPERIMENTAL	DIFFERENCE
$[1\ \bar{1}\ \bar{1}]$ and $[\bar{3}\ 1\ \bar{1}]$	139.5	138.0	1.5

After indexing the pattern, computer program determines the indices of zone axis for each zone.

It is mentioned in literature that the major growth direction for the primary dendrites in Sn-Bi alloys is [1 1 0], $[1\ \bar{1}\ 0]$ and the minor growth direction is [0 0 1]. Next, an attempt was made to determine whether the growth direction in Sn-Bi samples in the present study are [1 1 0], $[1\ \bar{1}\ 0]$ and [0 0 1]. These reported growth directions were located on the stereographic projections. The location of these directions was determined on the stereographic net using the expected angles between zone axes and the growth directions. The method of

locating [1 1 0], [1 $\bar{1}$ 0] and [0 0 1] on the stereographic projections is explained in section 4.6.

The angle between zone axes and the center of the stereographic projection (which represents the growth direction of the ingot) for samples are given in Table 1.

Table 1- Angle between indexed zone axes and ingot axis.

Sample No.	Zone Axes	Angle with Center (deg.)
33	[$\bar{1}$ 1 0]	86
	[3 1 $\bar{3}$]	88
36	[1 $\bar{1}$ $\bar{1}$]	76
	[$\bar{3}$ 1 $\bar{1}$]	84
2.1	[3 $\bar{1}$ 1]	80
	[1 $\bar{1}$ 1]	87
3.1	[$\bar{1}$ 0 $\bar{3}$]	77
	[1 1 0]	82
4.3	[1 3 $\bar{3}$]	80
	[0 1 $\bar{1}$]	80
6.3	[$\bar{1}$ 1 $\bar{1}$]	80
	[$\bar{1}$ $\bar{1}$ 1]	84
12.4	[$\bar{2}$ 1 0]	82
	[$\bar{1}$ 2 0]	81

The angle in column 3 of table 1 was measured to check if these angles match with the angles calculated between the zone axes and one of the reported growth direction. If these angles matched with, it would mean that the ingot axis coincides with the reported growth direction.

The growth direction for each of the sample was determined by measuring the angle between the center and the reported growth direction. The growth direction for each of the sample is being given in Table 2.

Table2 - Measured angles between ingot axis and the reported growth directions.

Sample No.	Angle between ingot axis and reported growth direction		
	[1 1 0]	[1 $\bar{1}$ 0]	[0 0 1]
This work			
33	46	76	40
36	48	46	58
Sampath's work			
2.1	54	66	50
3.1	82	40	78
6.3	76	78	32
12.4	88	90	8

From table no. 2 it can be seen that the observed growth direction in samples used in the present study is not the same as reported in the literature. It is observed from the results, that the [110] direction is not coinciding with the ingot axis and the angular difference between [110] and ingot axis is observed to be 40 to 88 degrees. The other growth directions which were reported in the literature were also considered and the angular difference in case of [1 $\bar{1}$ 0] was found to be 40 to 90 degree. When the growth direction was considered as [0 0 1] the angular difference was found to vary in the range of 8 degree to 78 degree.

In the present work, we used 20 mA and 30 kV X-ray beam and due to some technical limitations the voltage of the X-ray tube could not be increased. The LBR patterns obtained at this voltage were lighter and accurate determination of the x and y coordinates was difficult. So, before concluding anything about the orientation of the alloys, which we used, it would be important to get the LBR pattern at higher voltage and indexed for determining the growth direction of primary dendrites in the alloys. It is expected that at higher voltage the Laue back reflection pattern will be sharper and the measurement of coordinates in the pattern will be more accurate. Since the computer program takes coordinates of spots as the input data, the more the accuracy in measurement of coordinates, more accurate will be the determination of orientation.

The orientation of the directionally solidified Sn-Bi alloys can be separately determined using transmission electron microscopy also.

CHAPTER 6

CONCLUSIONS

1. A Computer code in 'C' language has been developed for indexing of Laue back reflection patterns from body centered tetragonal metals and alloys. The computer program assigns indices to the spots and their zone axes.

2. The computer program has been used for indexing of two Sn-Bi samples from directionally solidified ingots. The crystallographic orientations of both the samples were determined, using the code.

3. The directionally solidified Sn-Bi alloys were electropolished at room temperature of about 20 degrees Celsius. A suitable combination of voltage and current for electrolytic polishing was found to be 45 volt and 250-300 mA per sq. cm . The time for polishing was found to be 45 seconds for getting a good polished surface. The dendritic microstructure was visible under an optical microscope after electropolishing.

4. The growth direction of primary dendrites in Sn-Bi alloys is within a few degrees of the axis of the ingot. On the basis of the crystallographic orientation as determined using Laue back reflection patterns from the directionally solidified Sn-Bi alloys the angle between the axis of ingot and the expected growth directions, of primary dendrites [1 1 0], [1 $\bar{1}$ 0] and [0 0 1] as reported in literature, ranges from 40 degree to 76 degree.

5. The growth direction reported in literature for primary dendrites in dilute alloys of Bi in Sn have not been verified in the present work.

CHAPTER 7

SCOPE FOR FUTURE WORK

The present work can be modified or extended in two area,

1. In the experimental work for getting a better Laue pattern
2. Modification of computer program to make it more versatile.
3. The orientation of the alloys can be verified by transmission electron microscopy.

1. The samples were exposed to white radiation at 20 mA and 30 kV (maximum permissible on the machine) and it had been found that at this voltage the patterns were weak and the spots are lighter even after exposure times of upto 14 hrs. At the same time due to low voltage the mark from Pb marks in the film cassette, which were expected to come on the pattern did not come. So, the measurement of x and y coordinates and determination of film center were ambiguous. It is expected that at higher voltage the pattern will be more intense and take less time. The determination of coordinates will also be more accurate. Better accuracy in the measurement of coordinates of spots and location of center better will improve the output. So the sample should be exposed to X-rays at 40 kV. Laue patterns from more samples and from different grains of a sample need to be taken and indexed.
2. The present program has been written for indexing of body centered tetragonal system. This program can be easily modified for indexing of any crystal system. An arrangement in program can be made so that program will ask the system to be indexed and depending upon the system to be indexed, the structure factor condition will be applied and the crystallographic formulae for that system will be used. Since the angle calculation and generation of possible set of Miller indices have been

made in separate function the above change can be carried out without changing the algorithm used in this program.

3. The orientation of the directionally solidified alloys of Sn-Bi can be determined with the help of transmission microscopy for the confirmation of growth direction of the alloys.

These modifications mentioned above are not carried out in this project due to the time limitation.

CHAPTER 8

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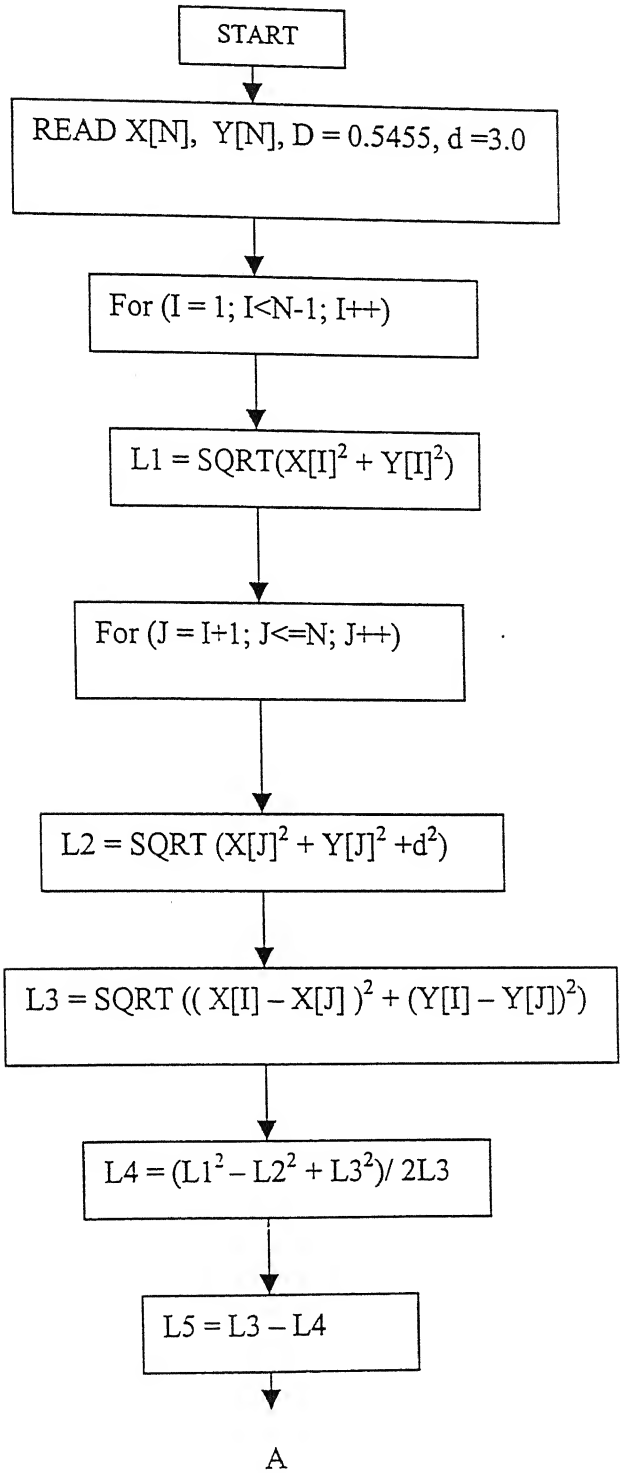
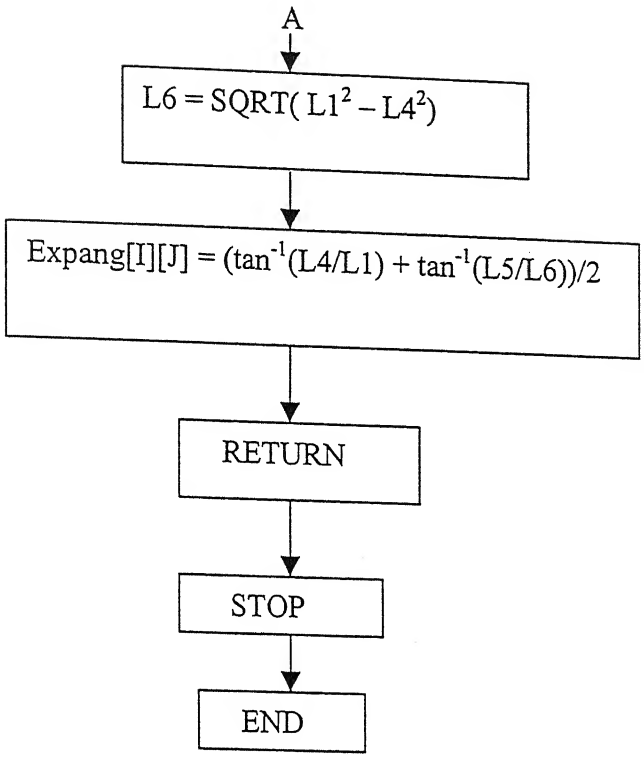
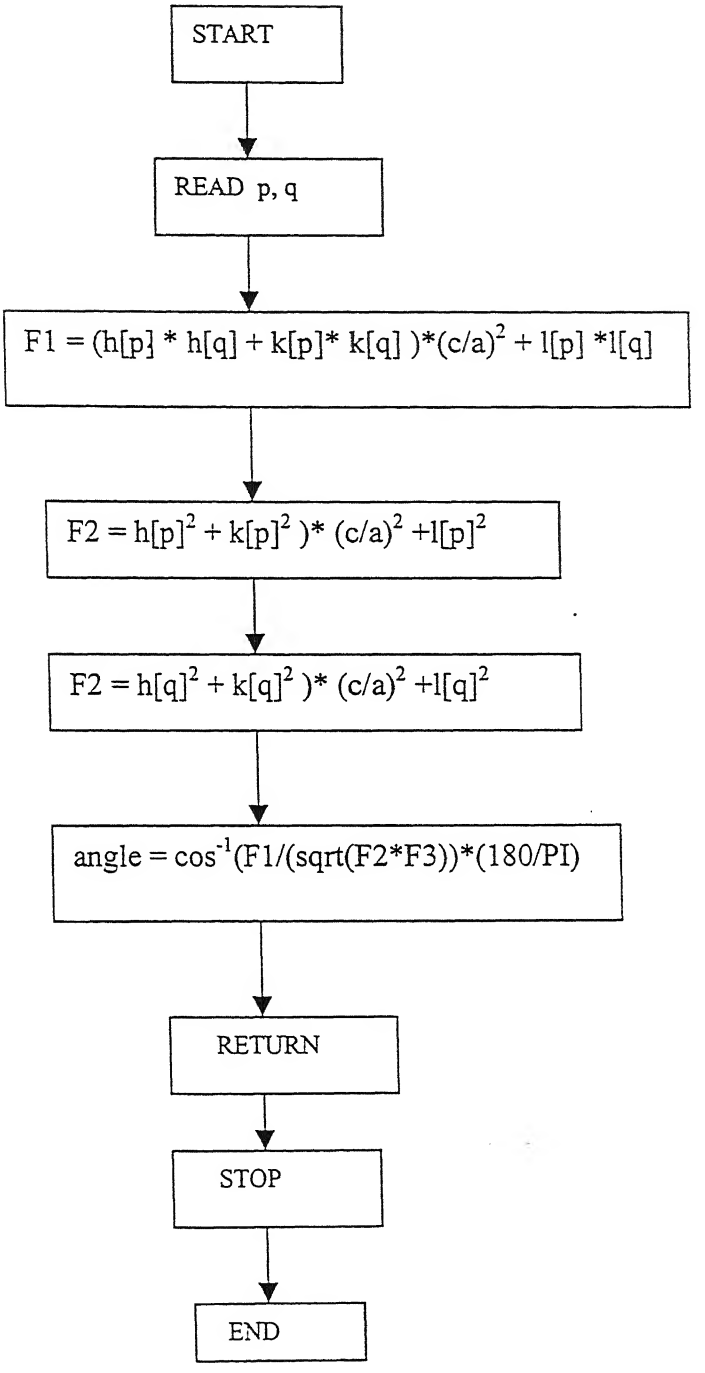
FLOW CAHRT OF FUNCTION EXPANG

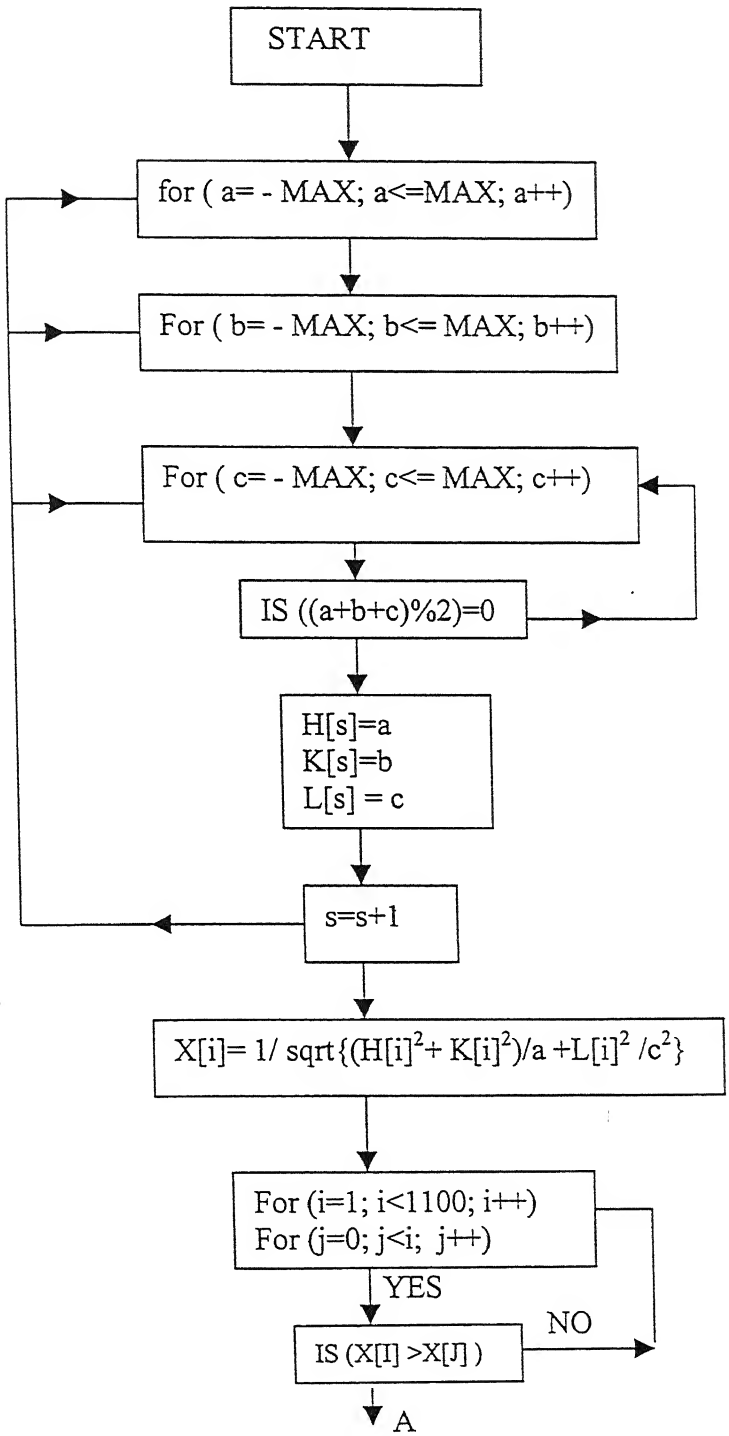


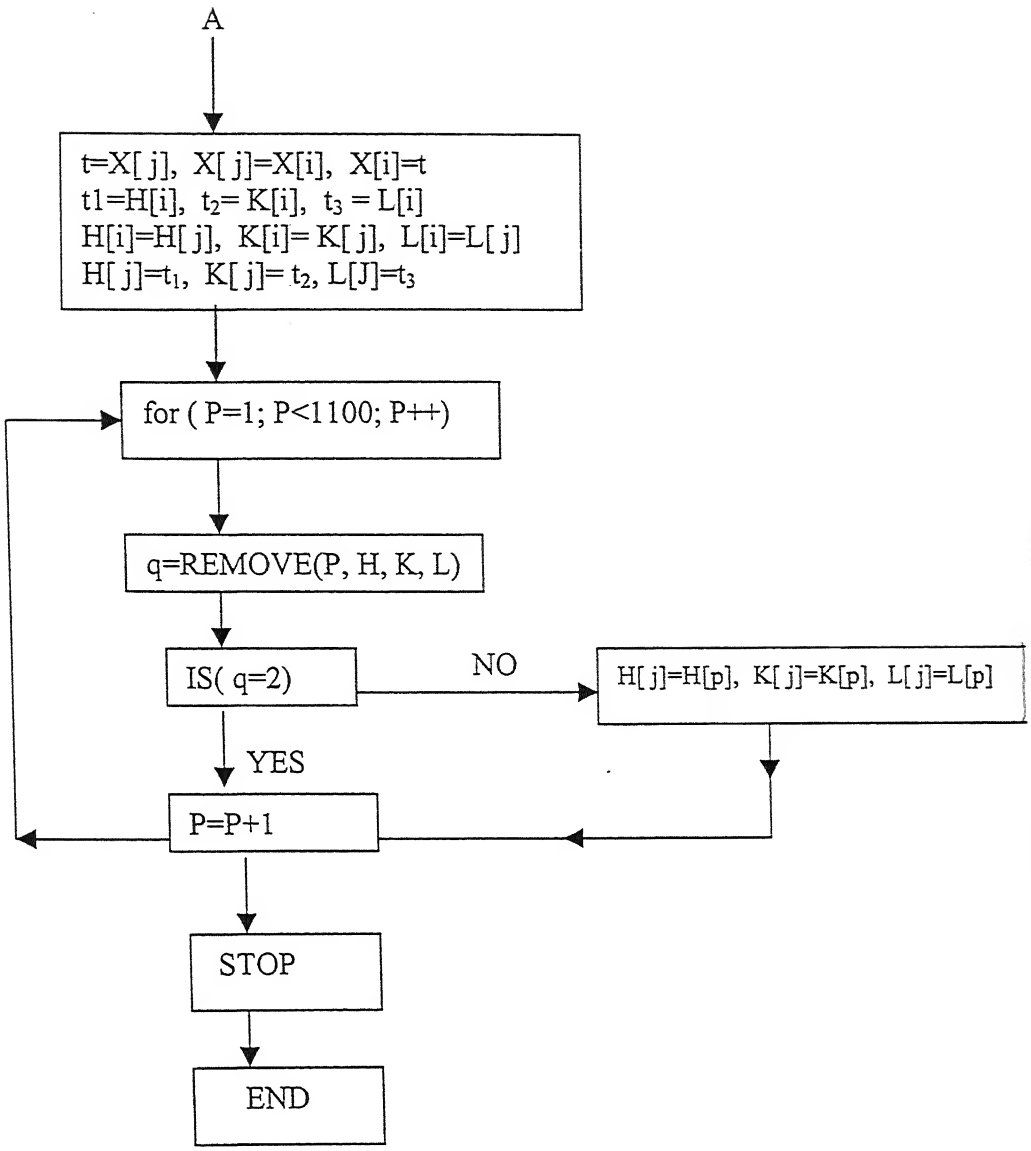

A
L6 = SQRT(L1^2 – L4^2)
Expang[I][J] = (tan^-1(L4/L1) + tan^-1(L5/L6))/2
RETURN
STOP
END

FLOW CHART OF FUNCTION TANGLE

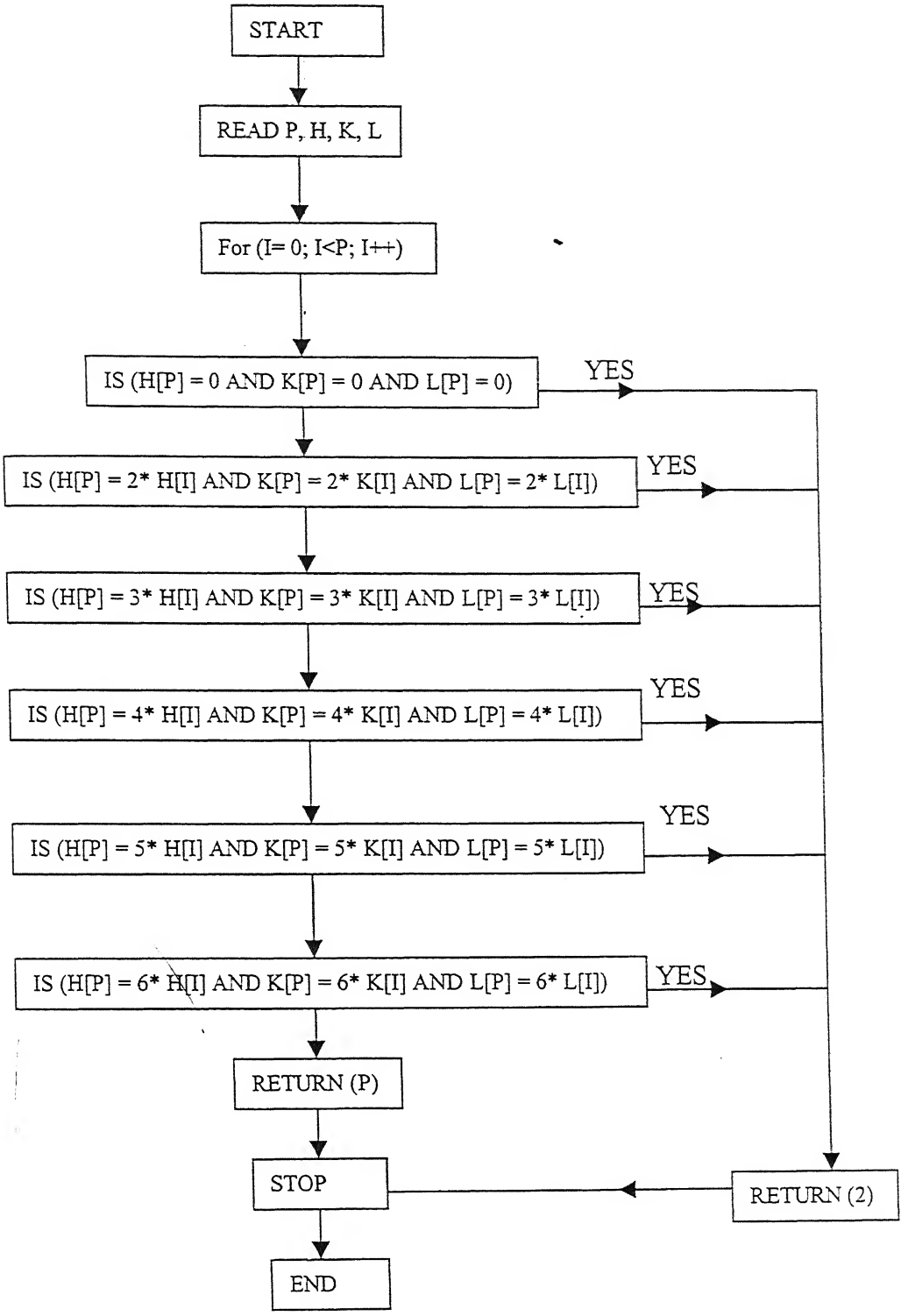


FLOW CHART OF FUNCTION INDICES

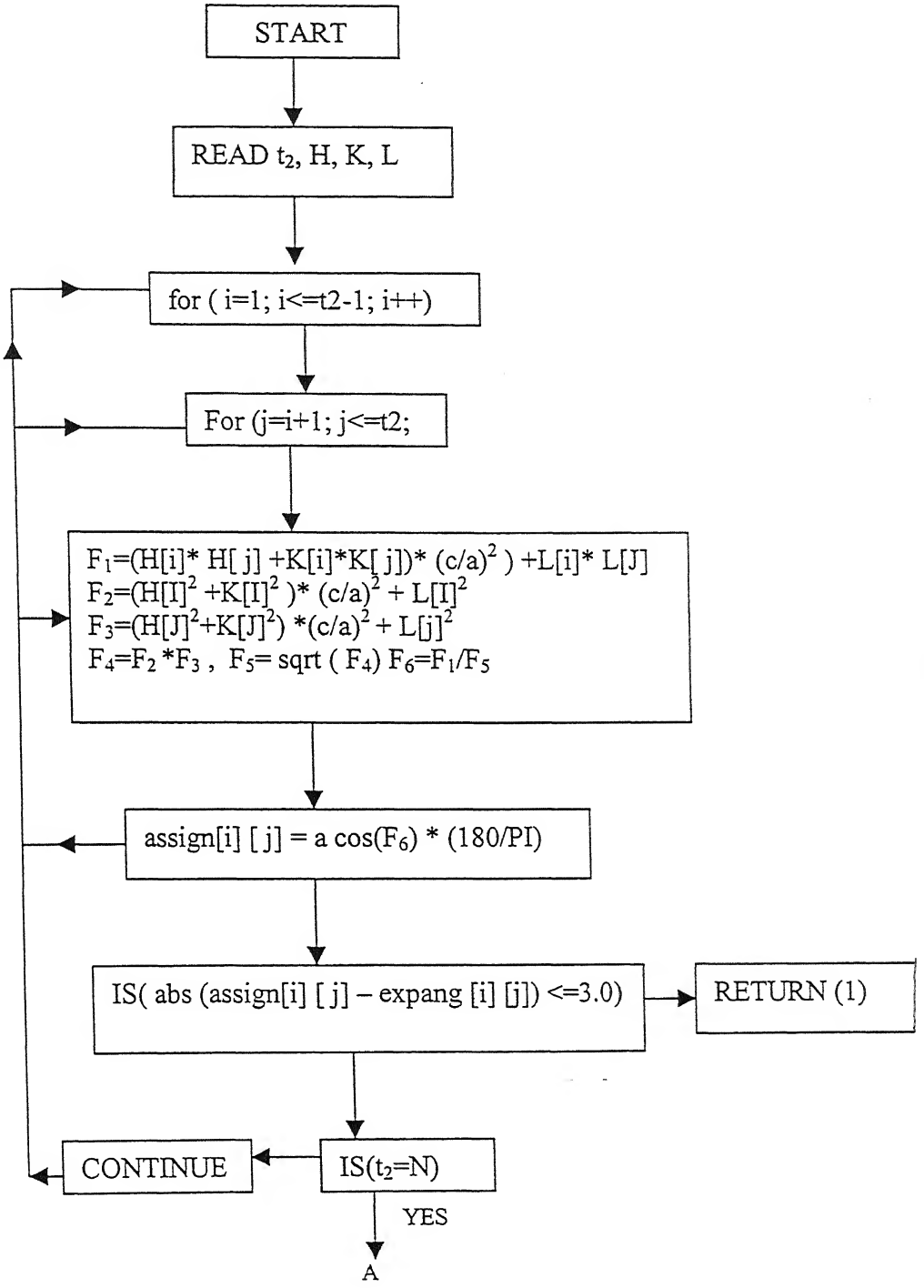


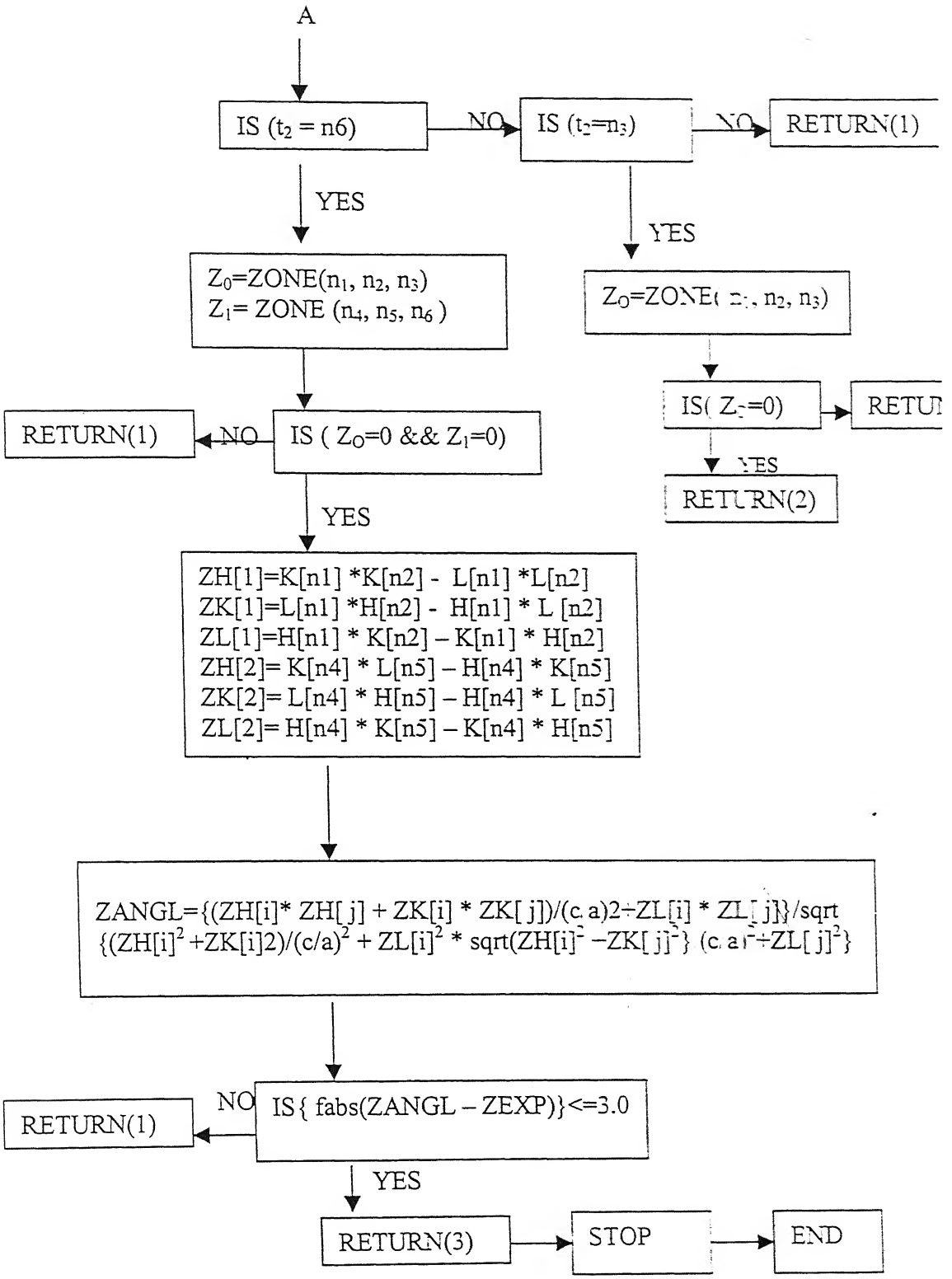

A
t=X[j], X[j]=X[i], X[i]=t
t1=H[i], t2= K[i], t3 = L[i]
H[i]=H[j], K[i]= K[j], L[i]=L[j]
H[j]=t1, K[j]= t2, L[J]=t3
for (P=1; P<1100; P++)
q=REMOVE(P, H, K, L)
IS(q=2)
NO
H[j]=H[p], K[j]=K[p], L[j]=L[p]
YES
P=P+1
STOP
END

FLOW CHART OF FUNCTION REMOVE

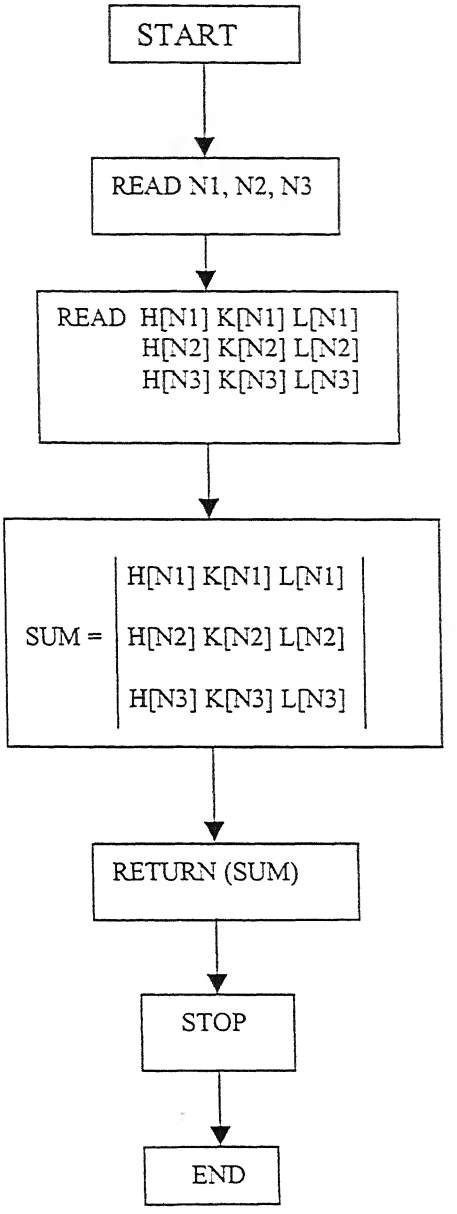


FLOW CHART OF FUNCTION CHECK

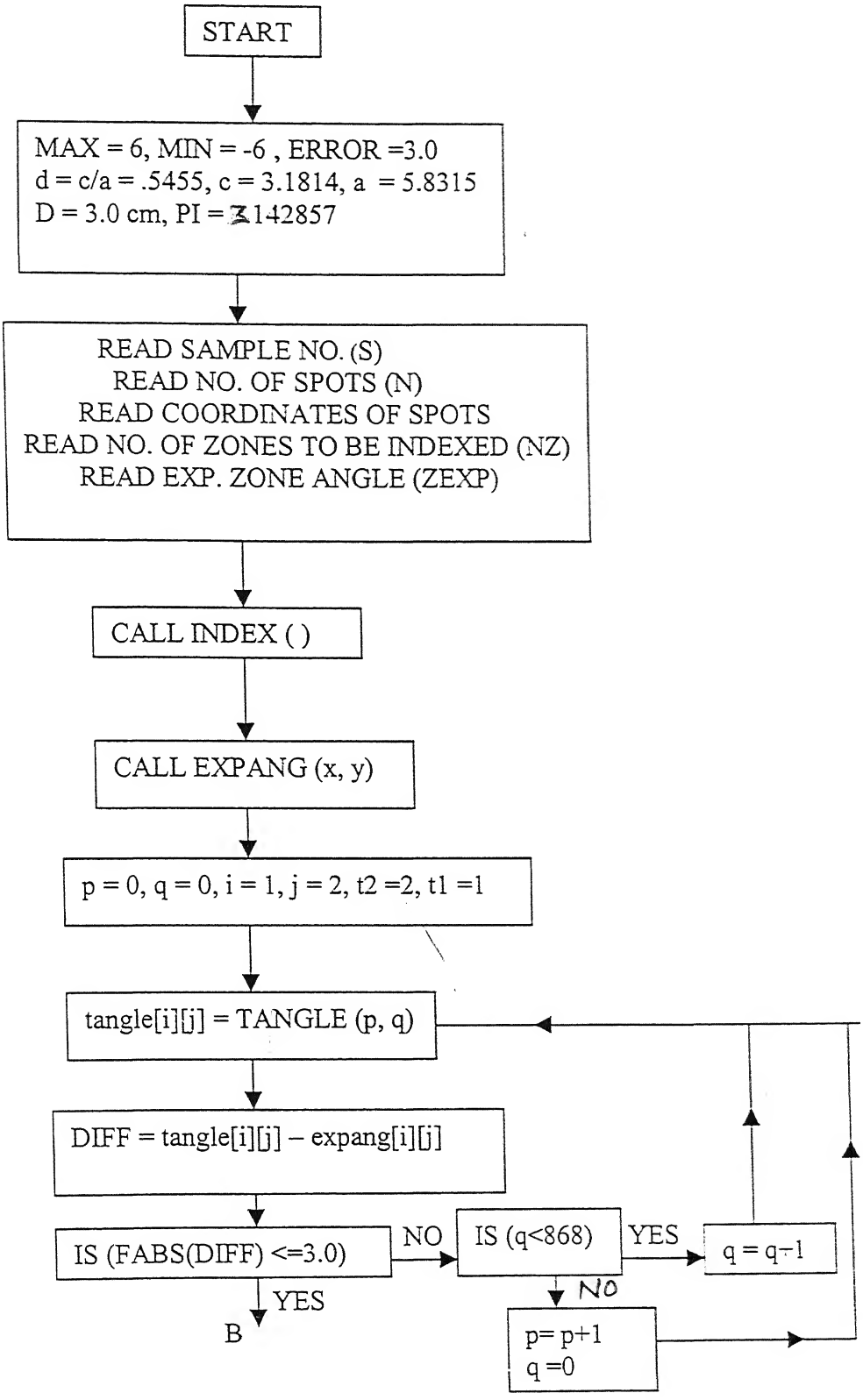


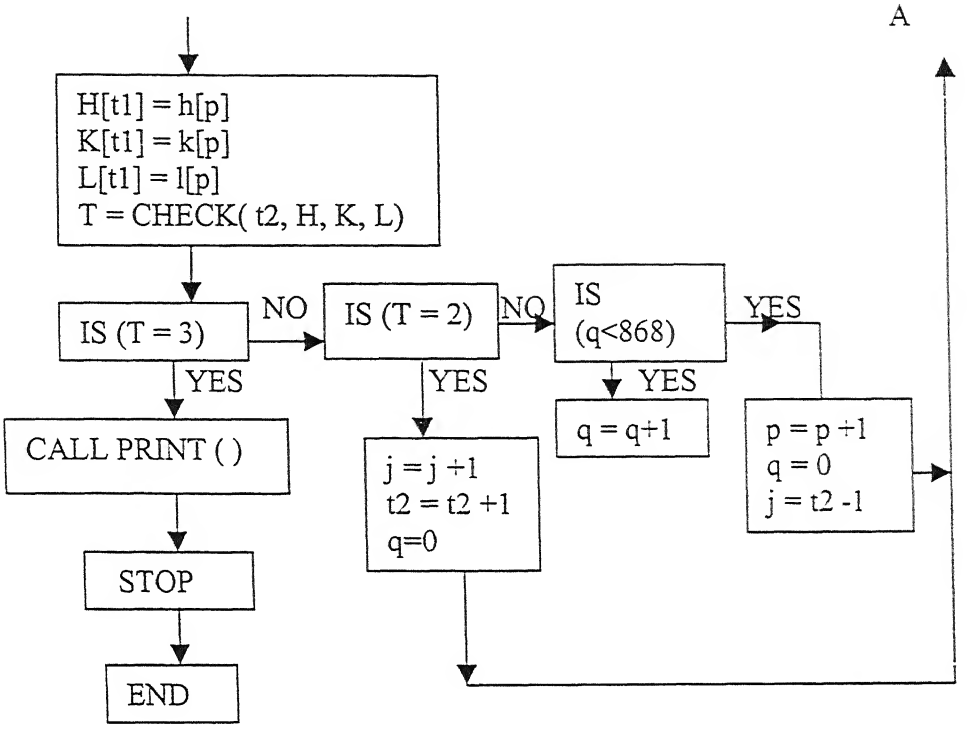

A
IS (t_2 = n6)
NO
IS (t_2=n_3)
NO
RETURN(1)
YES
YES
Z_0=ZONE(n_1, n_2, n_3)
Z_1= ZONE (n_4, n_5, n_6)
Z_0=ZONE(n_1, n_2, n_3)
IS(Z_0=0)
RETU
YES
RETURN(2)
RETURN(1)
NO
IS (Z_0=0 && Z_1=0)
YES
ZH[1]=K[n1] *K[n2] - L[n1] *L[n2]
ZK[1]=L[n1] *H[n2] - H[n1] * L [n2]
ZL[1]=H[n1] * K[n2] – K[n1] * H[n2]
ZH[2]= K[n4] * L[n5] – H[n4] * K[n5]
ZK[2]= L[n4] * H[n5] – H[n4] * L [n5]
ZL[2]= H[n4] * K[n5] – K[n4] * H[n5]
ZANGL={(ZH[i]* ZH[j] + ZK[i] * ZK[j])/(c a)2÷ZL[i] * ZL[j]}/sqrt
{(ZH[i]^2 +ZK[i]2)/(c/a)^2 + ZL[i]^2 * sqrt(ZH[i]^2 –ZK[j]^2} (c a)^2÷ZL[j]^2}
RETURN(1)
NO
IS{ fabs(ZANGL – ZEXP)}<=3.0
YES
RETURN(3)
STOP
END

FLOW CHART OF FUNCTION ZONE



FLOW CHART OF MAIN PROGRAM

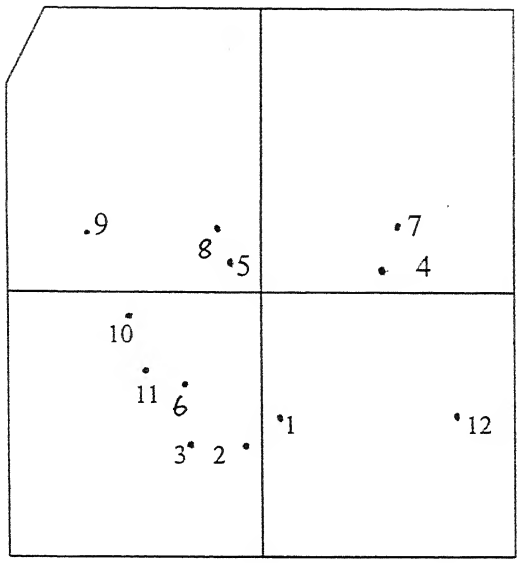



H[t1] = h[p]
K[t1] = k[p]
L[t1] = l[p]
T = CHECK(t2, H, K, L)
IS (T = 3)
NO
IS (T = 2)
NO
IS
(q<868)
YES
YES
YES
YES
CALL PRINT ( )
STOP
END
j = j +1
t2 = t2 +1
q=0
q = q+1
p = p +1
q = 0
j = t2 -1

APPENDIX B

Traces of patterns

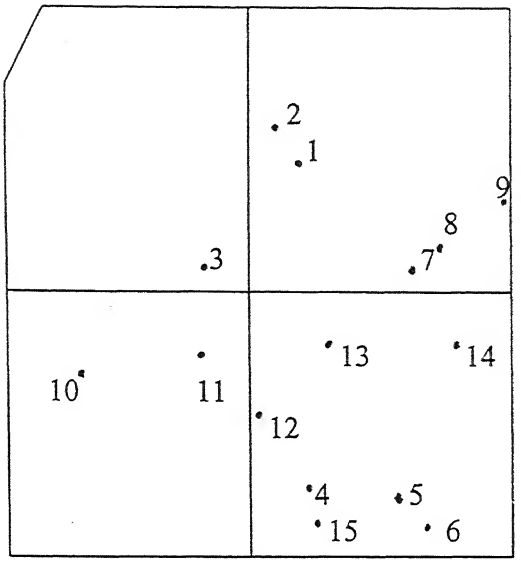
SAMPLE NO. 33



SPOT NO.	X (cm)	Y (cm)
1	0.40	- 1.50
2	- 0.30	- 1.80
3	- 0.80	- 1.80
4	1.80	0.10
5	- 0.60	0.70
6	- 0.80	—1.00
7	2.0	0.70
8	- 0.7	1 .00
9	- 2.6	1.30
10	- 1.9	- 0.50
11	- 1.7	- 1.20
12	3.2	-1.5

Traces of pattern

SAMPLE NO. 36



SPOT	X (cm)	Y (cm)
1	1.00	3.00
2	0.85	3.40
3	-0.60	0.60
4	0.80	-2.60
5	2.60	-3.00
6	2.80	-3.80
7	1.90	0.70
8	2.20	0.90
9	4.00	1.60
10	-2.70	-1.10
11	-0.70	-0.90
12	0.10	-1.20
13	1.40	-0.70
14	3.00	-0.80
15	0.90	-3.20

APPENDIX C

Computer outputs

SAMPLE NUMBER 33

NUMBER OF SPOTS INDEXED = 6

COORDINATES OF SPOTS

0.40	-1.50
-0.3	-1.80
-0.80	-1.80
1.80	0.10
-0.60	0.70
-0.80	–1.00

ERROR LIMIT IS 3.0 DEGREE

POINTS	EXPERIMENTAL ANGLE (1)	THEORETICAL ANGLE (2)	DIFFERENCE (1 – 2)
(1, 2)	6.250815	6.121440	0.129375
(1, 3)	10.076114	8.956684	1.119430
(1, 4)	17.733395	15.527786	2.205609
(1, 5)	20.377829	19.784938	0.592891
(1, 6)	10.933287	8.239948	2.693339
(2, 3)	3.996808	2.835245	1.161564
(2, 4)	22.876343	21.117217	1.759126
(2, 5)	20.314559	20.673947	0.359389
(2, 6)	6.901205	4.048949	2.852256
(3, 4)	25.514661	23.791948	1.722713

(3, 5)	19.653779	21.643568	1.989789
(3, 6)	4.389957	4.230261	0.159696
(4, 5)	20.020920	18.299415	1.721505
(4, 6)	23.022955	21.202540	1.820415
(5, 6)	15.355325	17.413307	2.057982

INDEX SEARCH IS BETWEEN (6 6 6) AND ($\bar{6}$ $\bar{6}$ $\bar{6}$)

INDICES OF SPOT [1] IS →	1	1	2
INDICES OF SPOT [2] IS →	4	4	6
INDICES OF SPOT [3] IS →	3	3	4
INDICES OF SPOT [4] IS →	1	0	3
INDICES OF SPOT [5] IS →	1	0	1
INDICES OF SPOT [6] IS →	4	3	5

Spot 1, 2 and 3 belongs to zone [$\bar{1}$ 1 0]

Spot 3, 5 and 6 belongs to zone [3 1 $\bar{3}$]

ZONE	ZONE ANGLE		
	THEORETICAL	EXPERIMENTAL	DIFFERENCE
[$\bar{1}$ 1 0] and [3 1 $\bar{3}$]	113.4	112.0	1.4

Computer output 2

SAMPLE NUMBER 36

NUMBER OF SPOTS INDEXED = 6

COORDINATES OF SPOTS

1.0	3.00
0.85	3.40
-0.60	0.60
0.80	-2.60
2.60	-3.00
2.80	-3.80

ERROR LIMIT IS 3.0 DEGREE

POINTS	EXPERIMENTAL ANGLE (1)	THEORETICAL ANGLE (2)	DIFFERENCE (1 – 2)
(1, 2)	4.644451	2.979718	1.664734
(1, 3)	15.958081	16.320074	0.391993
(1, 4)	37.097812	37.082374	0.015438
(1, 5)	38.501190	36.045744	2.455446
(1, 6)	41.461314	40.931708	0.529606
(2, 3)	15.866015	18.166131	2.300117
(2, 4)	38.116955	40.062091	1.945136
(2, 5)	39.743529	38.944547	0.798981
(2, 6)	42.697589	43.776160	1.078571
(3, 4)	28.032251	30.530835	2.498584
(3, 5)	34.044391	33.484346	0.560045
(3, 6)	36.307810	38.961372	2.653563
(4, 5)	10.737547	8.297525	2.440021
(4, 6)	12.297384	11.825073	0.472311
(5, 6)	7.935074	5.477026	2.458048

INDEX SEARCH IS BETWEEN (6 6 6) AND ($\bar{6}$ $\bar{6}$ $\bar{6}$)

INDICES OF SPOT [1] IS → $\bar{4}$ $\bar{5}$ 1

INDICES OF SPOT [2] IS → $\bar{5}$ $\bar{6}$ 1

INDICES OF SPOT [3] IS → $\bar{2}$ $\bar{5}$ 1

INDICES OF SPOT [4] IS → $\bar{1}$ $\bar{3}$ 2

INDICES OF SPOT [5] IS → $\bar{3}$ $\bar{5}$ 4

INDICES OF SPOT [6] IS → $\bar{2}$ $\bar{3}$ 3

Spot 1, 2 and 4 belongs to zone [1 $\bar{1}$ $\bar{1}$]

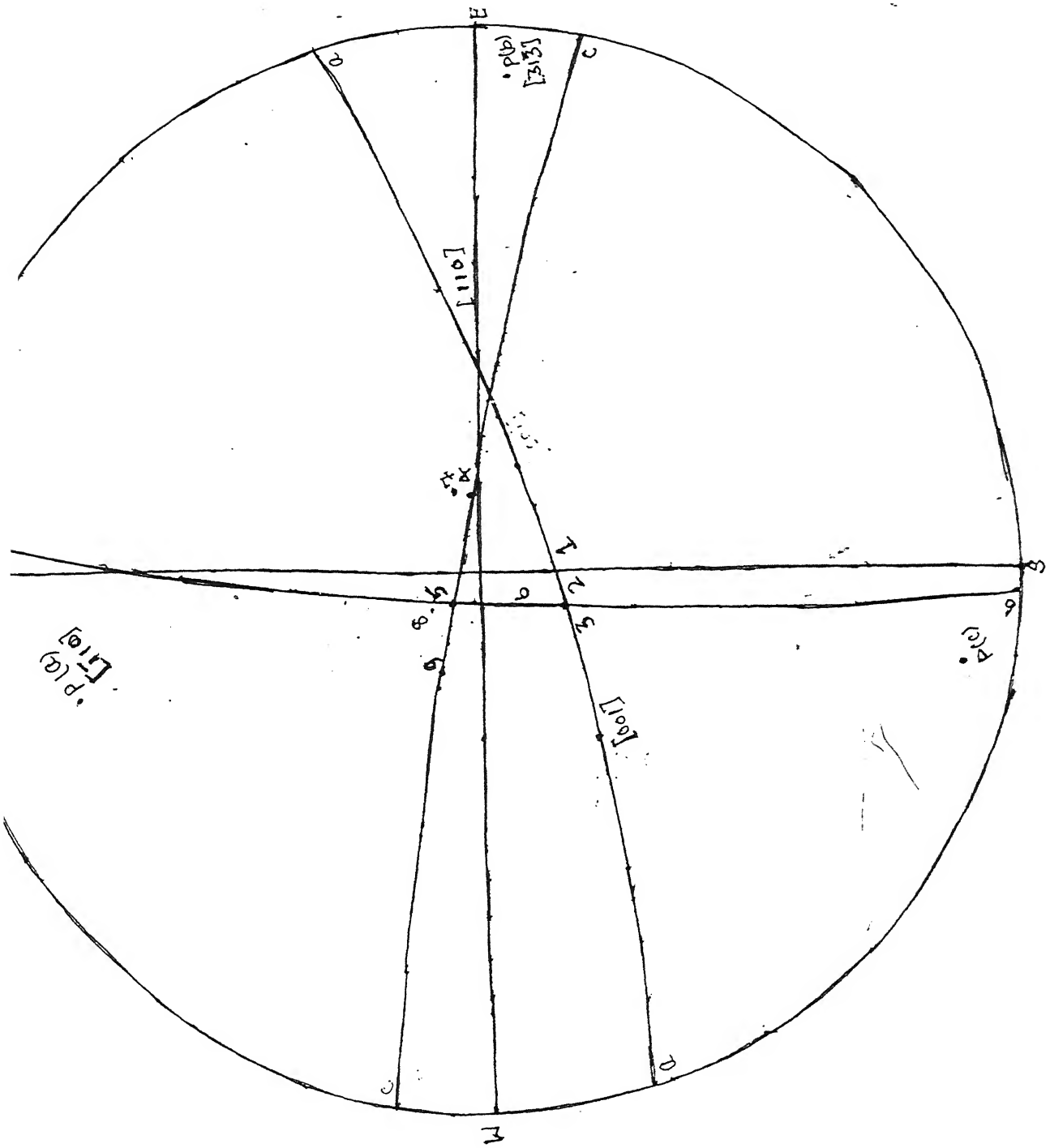
Spot 3, 5 and 6 belongs to zone [$\bar{3}$ 1 $\bar{1}$]

ZONE	ZONE ANGLE		
	THEORETICAL	EXPERIMENTAL	DIFFERENCE
[1 $\bar{1}$ $\bar{1}$] and [$\bar{3}$ 1 $\bar{1}$]	139.5	138.0	1.5

APPENDIX D

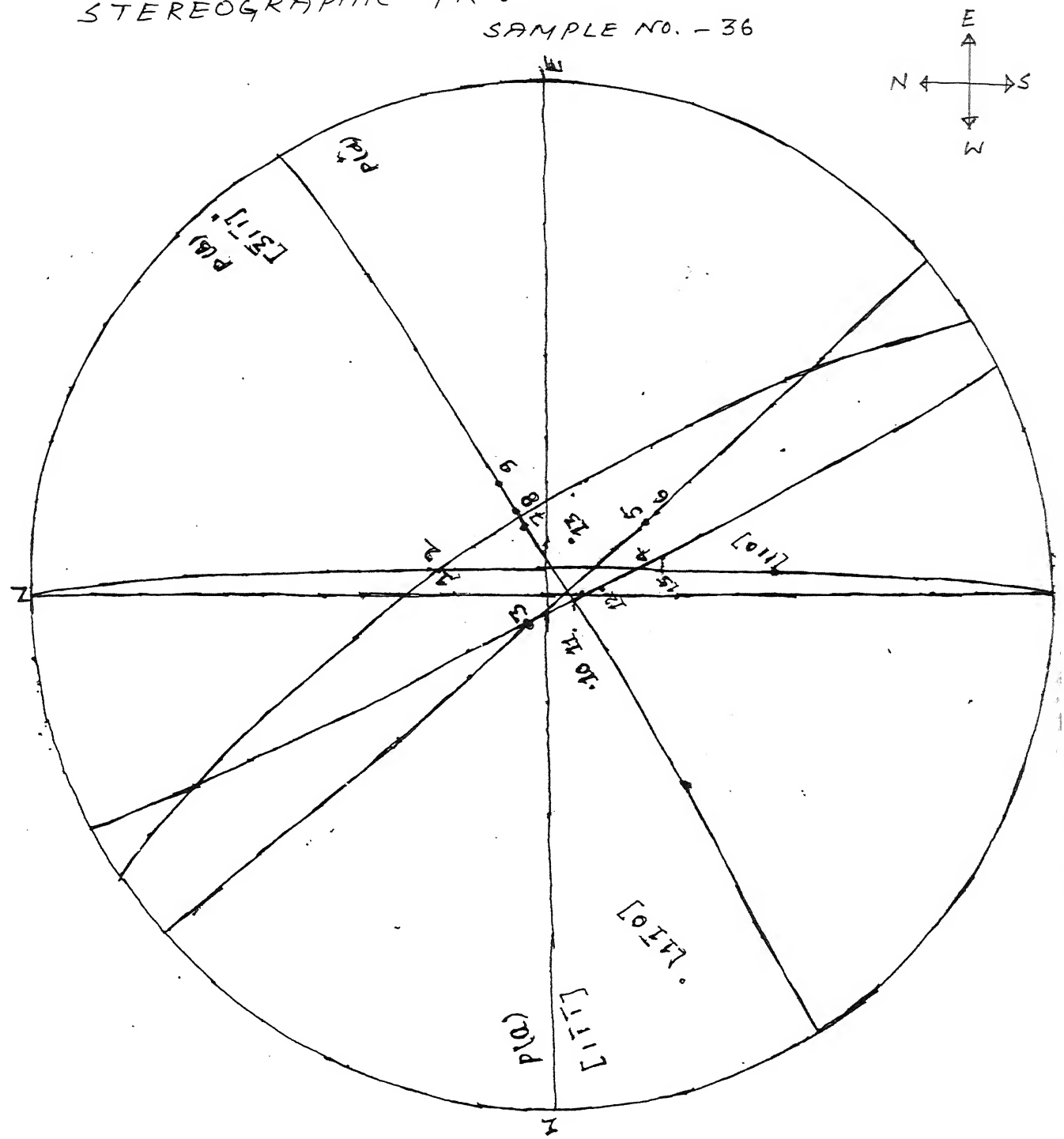
STEREOGRAPHIC PROJECTION

SAMPLE NO. - 33



STEREOGRAPHIC PROJECTIONS

SAMPLE NO. – 36



APPENDIX E

LIST OF VARIABLES

a	:	Lattice parameter
c	:	Lattice parameter
d	:	Separation of film from the sample (3.0cm)
D	:	c/a ratio (0.5455)
N	:	Number of spots to be indexed
S	:	Sample number
MAX	:	Maximum limit of Miller indices
MIN	:	Minimum limit of Miller indices
NZ	:	Number of zones to be considered
n1, n2, n3, n4, n5, n6, n7, n8, n9	:	Spots number
h[], k[], l[]	:	Arrays for storing Miller indices from which spots will be indexed
x[], y[]	:	Coordinates of spots
H[], K[], L[]	:	Arrays of Miller indices in which final indices will be stored after searching
ZH[], ZK[], ZL[]	:	Arrays for storing indices for zone axes
i, j, k ,l , t1, t2, t3, t4	:	Counter
expang[i][j]	:	Experimental angles are stored in this array calculated using their coordinates, i and j are the spots number
ZEXP[]	:	Arrays for storing experimental zone angle measured from stereographic projection

ZANGL[]	:	Arrays for storing theoretical zone angle calculated from crystallographic expression for body centered tetragonal system after assigning indices to spots.
tangle[i][j]	:	Theoretical angle between spots calculated from crystallographic expression after assigning indices to spot, i and j are the spot number
diff.	:	Difference between theoretical and experimental interplanar angle
i1, j1, p1, q1	:	Counter
A[], B[], C[]	:	Arrays to store rejected Miller indices after rejecting all the indices which are scalar multiple of some previous indices
x1[]	:	Arrays to store the interplanar spacing for Miller indices set
angle	:	Angle between spots calculated after assigning indices to spots. This angle is returned from function TANGLE( ) to the main program.
assign[i][j]	:	Angle between spot i and j calculated using the x and y coordinates of spot. This angle is calculated in function CHECK( ) to check the mutual angles difference.
Zdiff	:	Difference in experimental and theoretical zone angles.
Sum	:	The value of determinant, which is calculated for zone checking.

N1, N2, N3	:	Spots number of one zone under checking for zone condition
Z0, Z1, Z2	:	Stores the value of determinant of three spots of one zone
Store[]	:	Arrays to store Miller indices, which have already been assigned to spots so that same Miller indices can not be assigned to two spots.
T	:	Variable which takes value of CHECK(t2, H, K, L) and depending on the value of T the main program starts searching for indices.